Proceedings of the 14th
KACC, Octorber 1999

Multivariable Nonlinear Model Predictive Control of a Continuous Styrene Polymerization Reactor

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Abstract

Model predictive control algorithm requires a relevant model of the system to be controlled. Unfortunately, the first principle model describing a polymerization reaction system has a large number of parameters to be estimated. Thus there is a need for the identification and control of a polymerization reactor system by using available input-output data. In this work, the polynomial auto-regressive moving average (ARMA) models are employed as the input-output model and combined into the nonlinear model predictive control algorithm based on the successive linearization method.

Simulations are conducted to identify the continuous styrene polymerization reactor system. The input variables are the jacket inlet temperature and the feed flow rate whereas the output variables are the monomer conversion and the weight-average molecular weight.

The polynomial ARMA models obtained by the system identification are used to control the monomer conversion and the weight-average molecular weight in a continuous styrene polymerization reactor. It is demonstrated that the nonlinear model predictive controller based on the polynomial ARMA model tracks the step changes in the setpoint satisfactorily. In conclusion, the polynomial ARMA model is proven effective in controlling the continuous styrene polymerization reactor.

1. Introduction

A polymerization reaction system is subject to a complex reaction mechanism and shows highly nonlinear features. Thus, the first principles model of a polymerization reactor may contain a large number of kinetic parameters and nonlinear terms. These parameters are neither readily found from the literature nor easily determined by experiments. As an alternative to the first principles model, it may be advantageous to use a model structure whose parameters can be identified from input-output data. Since input-output models can effectively describe a nonlinear system, a number of those models have been employed for the construction of model-based controllers, such as the model predictive controller (MPC).

A wide variety of nonlinear input-output models have been proposed for use in identification and control of chemical processes. Fruzzetti *et al.* employed the Hammerstein model in a nonlinear MPC scheme for the pH control of a chemical reactor and for the control of a binary distillation column. Although the Hammerstein model is convenient for the controller design and also for handling the process noise, it is incapable of modeling systems that exhibit output multiplicities as displayed by various chemical systems.

Maner and Doyle III used the autoregressive plus Volterra model in order to identify a continuous methyl methacrylate (MMA) polymerization reactor and implemented the model in the control of polymer properties using a nonlinear MPC. Hernandez and Arkun[5] identified single-input-single-output (SISO) polynomial ARMA model for a CSTR in which first-order exothermic reaction occurred. The authors examined the steady-state multiplicity by using the model and proposed a nonlinear MPC scheme based on the polynomial ARMA model.

The main advantage of polynomial ARMA models is that it can describe the process nonlinearities as polynomial nonlinearities and can greatly simplify the estimation of the parameters from the input-output data. Moreover, polynomial ARMA models are superior to Volterra series models in the sense that the number of parameters needed to approximate a system is, in general, much less with polynomial models. However, the polynomial ARMA models have some problems that must be solved for application to nonlinear MPC. The most severe problem is that the objective function becomes a higher order function of the input variables as prediction horizon is increased. A higher order objective function could be expected to lead to a less efficient and possibly unreliable solution using current nonlinear optimization techniques.

Garcia (1984) proposed nonlinear quadratic dynamic matrix control (NLQDMC) based on successive linearization of the nonlinear model. Lee and Ricker (1994) developed a computationally efficient nonlinear control algorithm by combining the main concepts of NLQDMC with the extended Kalman filter. Both algorithms find extensive industrial applications, mainly by virtue of mild computational requirement.

In this study, we perform the multi-input-multi-output (MIMO) identification for a continuous styrene polymerization reactor using the polynomial ARMA model by simulation. Then we design the nonlinear MPC based on the polynomial ARMA models by using the successive linearization method. Simulations are performed to control the conversion and the weight-average molecular weight in a continuous styrene polymerization reactor.

2. Identification

A multi-variable polynomial ARMA model, which is a simple case of the general nonlinear ARMA model, is defined as follows :

$$\begin{aligned} y_i(k) = y_{i0} &+ \sum_{l=1}^{q_y}\sum_{j=1}^{n_y}\theta_{l,j}^{1,i}y_l(k-j) + \sum_{l=1}^{q_u}\sum_{j=1}^{n_u}\theta_{l,j}^{2,i}u_l(k-j) \\ &+ \sum_{l=1}^{q_y}\sum_{j=1}^{n_y}\sum_{m=l}^{q_y}\sum_{n=1}^{j}\theta_{l,j,m,n}^{3,i}y_l(k-j)y_m(k-n) \\ &+ \sum_{l=1}^{q_u}\sum_{j=1}^{n_u}\sum_{m=l}^{q_u}\sum_{n=1}^{j}\theta_{l,j,m,n}^{4,i}u_l(k-j)u_m(k-n) \\ &+ \sum_{l=1}^{q_y}\sum_{j=1}^{n_y}\sum_{m=1}^{q_u}\sum_{n=1}^{n_u}\theta_{l,j,m,n}^{5,i}y_l(k-j)u_m(k-n)+\cdots \end{aligned} \quad (1)$$

where q_y and q_u denote the numbers of outputs and inputs, respectively, while n_y and n_u indicate the number of lags on the outputs and inputs, respectively. In this study, input variables are the jacket inlet temperature and the feed flow rate, whereas output variables are the monomer conversion and the weight average molecular weight.

The nonlinear order of the model and the number of lags on the inputs and outputs were empirically specified as $n = 2$, $n_u = 3$, and $n_y = 3$, respectively. The inputs and outputs were scaled as follows:

$$y_1 = X \quad , \quad y_2 = \left(M_w - M_{w\,\min}\right)/\left(M_{w\,\max} - M_{w\,\min}\right) \quad (2)$$

$$u_1 = \frac{T_{jin} - T_{jin\,\min}}{T_{jin\,\max} - T_{jin\,\min}} \quad , \quad u_2 = \frac{q_f - q_{f\,\min}}{q_{f\,\max} - q_{f\,\min}} \quad (3)$$

where X, M_w, T_{jin}, and q_f denote the monomer conversion, the weight average molecular weight, the jacket inlet temperature, and the feed flow rate, respectively. The scaling factors in eqs (2) and (3) are specified in Table 1.

Table 1 Scaling factors for input and output variables

Weight-average molecular weight (M_w, y_2)	$M_{w\,min}$	10000
	$M_{w\,max}$	60000
Jacket inlet temperature (T_{jin}, u_1)	$T_{jin\,min}$	55 °C
	$T_{jin\,max}$	85 °C
Feed flow rate (q_f, u_2)	$q_{f\,min}$	5 ml/min
	$q_{f\,max}$	30 ml/min

In order to perform the simulation study, we consider the mathematical model of a continuous stirred-tank reactor in which solution polymerization of styrene occurs. The reaction kinetics is assumed to follow the free radical polymerization mechanism including chain transfer reactions to both solvent and monomer.

One can derive the mass balance equations for the initiator, solvent, and monomer, respectively, as follows:

$$\begin{aligned}
\frac{d(IV)}{dt} &= q_f I_f - qI + (-k_d)IV \\
\frac{d(SV)}{dt} &= q_f S_f - qS + (-k_{trs} S G_0)V \\
\frac{d(MV)}{dt} &= q_f M_f - qM - \{2fk_d I + (k_p + k_{trm})MG_0\}V
\end{aligned} \tag{4}$$

For the first three moments of living and dead polymer concentrations, the mass balance equations can be expressed as

$$\begin{aligned}
\frac{d(G_0 V)}{dt} &= -qG_0 + (2fk_d I - k_{tc} G_0^2)V \\
\frac{d(G_1 V)}{dt} &= -qG_1 + \{2fk_d I + k_p M G_0 - k_{tc} G_0 G_1 \\
&\quad + (k_{trm} M + k_{trs} S)(G_0 - G_1)\}V \\
\frac{d(G_2 V)}{dt} &= -qG_2 + \{2fk_d I + k_p M(G_0 + 2G_1) - k_{tc} G_0 G_2 \\
&\quad + (k_{trm} M + k_{trs} S)(G_0 - G_2)\}V \\
\frac{d(F_0 V)}{dt} &= -qF_0 + \{0.5k_{tc} G_0^2 + (k_{trm} M + k_{trs} S)G_0\}V \\
\frac{d(F_1 V)}{dt} &= -qF_1 + \{(k_{tc} G_0 + k_{trm} M + k_{trs} S)G_1\}V \\
\frac{d(F_2 V)}{dt} &= -qF_2 + \{k_{tc}(G_0 G_2 + G_1^2) + (k_{trm} M + k_{trs} S)G_2\}V
\end{aligned} \tag{5}$$

The energy balances for the reactor and the jacket give rise to the following equation

$$\begin{aligned}
\frac{d((\rho C_p)_m V T_r)}{dt} &= (\rho C_p)_m q_f T_f - (\rho C_p)_m q T_r + (-\Delta H_p)k_p M G_0 V - UA(T_r - T_j) \\
\frac{d((\rho C_p)_c V_j T_j)}{dt} &= (\rho C_p)_c q_c T_{jin} - (\rho C_p)_c q_c T_{jout} + UA(T_r - T_j) + U_a A_a (T_a - T_j)
\end{aligned} \tag{6}$$

In order to take into account the gel effect, we use the empirical correlation suggested by Hamer *et al.*, which is listed in Table 2. The physical properties and kinetic parameters were taken from the literature and are also listed in Table 2.

Reference conditions of simulation study are summarized in Table 3. The regressors and model parameters were obtained by using a stepwise model building algorithm (Kortmann *et al.*, 1988). In this algorithm, we used both popular F-test and Akaike's information criterion as model verification methods. Since the F-test seems to have the disadvantage of being difficult to choose the proper levels of significance, Akaike's information criterion was used together in model identifications. The procedure of stepwise model building algorithm quits if the F-test shows that the model equation is not significant or if Akaike's information criterion gives a value greater than that for the previous model.

Table 2 Physico chemical data and the gel effect correlation used in the mathematical for styrene polymerization.

Parameters	Values	Reference
	Physical Properties	
ρ_m[g/l]	924.0-0.981 × (T-273.15)	Schuler and Suzhen (1985)
ρ_s[g/l]	885.5-0.955 × (T-273.15)	"
ρ_p[g/l]	1084.0-0.605 × (T-273.15)	Takamatsu *et al.* (1988)
	Rate Constants	
k_d[s^{-1}]	$1.58\times10^{15}\exp(-30780/RT)$	Duerksen *et al.* (1967)
k_p[l/mol/s]	$1.051\times10^{7}\exp(-7064/RT)$	"
k_{t0}[l/mol/s]	$1.255\times10^{9}\exp(-1680/RT)$	"
k_{trm}[l/mol/s]	$1.186\times10^{7}\exp(-11767/RT)$	Yoo and Rhee (1999)
k_{trs}[l/mol/s]	$3.148\times10^{9}\exp(-16264/RT)$	"
	Gel Effect Correlation	
$g_t(X,T) = \frac{k_t}{k_{t0}} = \exp[-2(AX + BX^2 + CX^3)]$ where $A = 2.57 - 5.05\times10^{-3}T$, $B = 9.56 - 1.76\times10^{-2}T$, $C = -3.03 - 7.85\times10^{-3}T$		Hamer *et al.* (1981)

Although a 3-level sequence is persistently exciting, the results of preliminary study revealed that a 4-level sequence was superior to a 3-level sequence. Hence, the input signals, u_1 and u_2, were drawn from a uniform distribution with 4 levels. Just like the pseudo random binary signals (PRBS), the pseudo random multi-level signals are persistently exciting for a system, but the latter have several additional features. The pseudo random multi-level signals include interior points of input region, are closer to white autocorrelation function, and excite nonlinear modes that PRBS cannot. Because of these features, we used the pseudo random multi-level input signals rather than the PRBS. The process white noise with zero mean and standard deviation of 0.02 was added to both the conversion and the molecular weight. The output data obtained by the mathematical model against the pseudo random multi-level

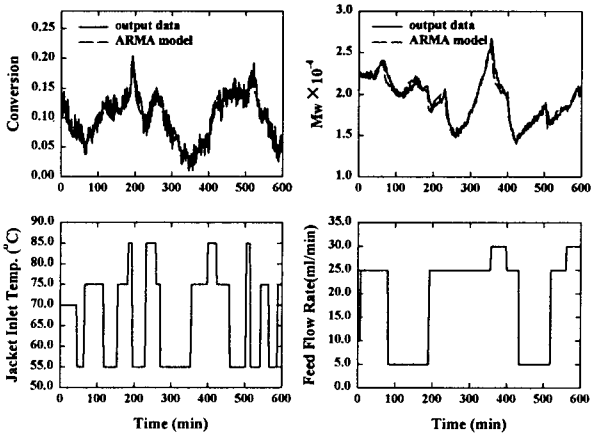

Figure 1 Output data obtained by the mathematical model against the pseudo random multi-level input signals with P_s=0.1 and 4 levels and the response of the identified polynomial ARMA model.

Table 3 Reference conditions for simulation study.

Initial Charge	Monomer(Styrene)	400 mL
	Solvent(Toulene)	400 mL
	Initiator(AIBN)	8g
Feed Concentration	Monomer	4.34 mol/L
	Solvent	4.70 mol/L
	Initiator	0.06 mol/L
Operating conditions	Reactor temperature	50-85 °C
	Feed flow rate	5-30 mL/min
Reactor volume (V)		1 L
Jacket volume (V_j)		0.8 L
Feed temperature (T_f)		20 °C
Ambient temperature (T_a)		20 °C
Initiator efficiency (f_i)		0.5
Heat of reaction for propagation (H_p)		74500 cal/mol
Thermal conductance		
UA		16 cal/sec/K
U_aA_a		3.2 cal/sec/K
Heating or cooling water flow rate (q_c)		2.5 L/min

input signals and the response of the identified polynomial ARMA model are shown in Figure 1. In this case, the sample time was 2 min and switching probability P_s, which represents the probability of input change at the end of any sampling interval, was assumed to be 0.1. The results of the identification are summarized in Table 4 (Na and Rhee, 1999).

Table 4 Simulation results of the identification using pseudo random multi-level input signals with P_s=0.1 and 4 levels.

	Conversion $y_1(k)$		Weight average molecular weight y_2(k)	
VAF	89.66%		95.3%	
	Para-meters	Regressors	Para-meters	Regressors
Models	0.02	1	-0.08	1
	0.20	y_1(k-1)	0.40	y_2(k-1)
	0.34	y_1(k-2)	-0.48	y_2(k-2) u_2(k-3)
	0.04	u_1(k-1) u_1(k-2)	0.59	y_2(k-2)
	-0.18	y_1(k-2) u_2(k-1)	0.15	y_1(k-3) u_2(k-3)
	0.32	y_1(k-3)	0.49	y_2(k-3)
	-0.02	y_2(k-1)	0.05	y_1(k-1)^2
	-0.09	y_1(k-1) u_1(k-1)	-0.22	u_1(k-1) u_1(k-2)
	-0.0025	u_1(k-2) u_1(k-3)	0.11	y_1(k-2) u_1(k-1)
			0.04	u_1(k-2)
			0.05	u_1(k-1) u_2(k-1)
			-0.19	y_2(k-2) u_1(k-2)
			-0.22	y_2(k-2) y_2(k-3)
			-0.22	y_2(k-1) y_2(k-2)
			-0.27	y_2(k-3)^2
			0.25	u_1(k-1)
			0.21	y_1(k-2)
			-0.03	u_2(k-1)^2
			0.03	u_2(k-1) u_2(k-2)
			0.16	u_1(k-1)u_1(k-3)

3. Controller design and simulation study

The objective function considered in this work is defined as follows:

$$J = \sum_{i=1}^{p} \left\{ \Lambda_y \left[y(k+i) - r(k+i) \right] \right\}^2 + \sum_{j=1}^{m} \left\{ \left[\Lambda_u \Delta u(k+j-1) \right] \right\}^2 \quad (7)$$

where Λ_y and Λ_u denote the weighting factors for the output and input, respectively, and $r(k+i)$ represents a reference value. The outputs are predicted by the identified ARMA model over the prediction horizon p. Unfortunately, however, eq (7) becomes a higher order function of the input sequence as p is increased because several output regressors are included in identified ARMA model. Therefore, the minimization of eq (7) for the input sequence is computationally more burdensome in the on-line control.

Based on local linear approximations of state/measurement equations computed at each sample time, a recursive state estimator providing the minimum-variance state estimates (known as the "extended Kalman Filter (EKF)") is derived. The same local approximation of the state equation is made linear with respect to the undecided control input moves by making linear approximations dual to those made for the EKF. Minimization of the weighted 2-norm of the tracking errors with various constraints can be solved via quadratic programming (Lee and Ricker, 1994).

Following equations are derived from state-space realization for a polynomial ARMA model by using delay coordinate method.

$$\begin{aligned} x(k) &= A \cdot x(k-1) + B \\ y(k) &= \begin{bmatrix} 1 & 0 & \cdots & \cdots & \cdots & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \end{bmatrix} \cdot x(k) \end{aligned} \quad (8)$$

The state vector in eq (8) is defined as follows:

$$\begin{aligned} &x(k) = \left[x_1(k) \;\; x_2(k) \;\; \cdots\cdots \;\; x_{14}(k) \right]^T \\ &\textit{where} \\ &x_1(k) = y_1(k),\; x_2(k) = y_2(k),\; \cdots,\; x_8(k) = y_2(k-3), \\ &x_9(k) = u_1(k-1), x_9(k) = u_2(k-1) \cdots, x_{14}(k) = u_2(k-3) \end{aligned} \quad (9)$$

The matrix A and B in eq (8) are derived as eq (10), whereas eq (8) is simply expressed by eq (11).

$$A = \begin{bmatrix} 0 & 0 & \cdots & 0 & & & \\ 0 & 0 & \cdots & 0 & & & \\ 1 & 0 & \cdots & 0 & & & \\ 0 & 1 & \cdots & 0 & & & \\ & & & \vdots & & & \\ & & & \vdots & & & \\ & & & & 0 & \cdots & 0 \\ & & & & 0 & \cdots & 0 \\ & & & & 1 & \cdots & 0 \\ & & & & & \ddots & \vdots \\ & & & & & & 1 \end{bmatrix}, \; B = \begin{bmatrix} p_1[x(k), u(k)] \\ p_2[x(k), u(k)] \\ 0 \\ 0 \\ \vdots \\ \vdots \\ u_1(k) \\ u_2(k) \\ 0 \\ \vdots \\ 0 \end{bmatrix} \quad (10)$$

$$\begin{aligned} x(k) &= F[x(k-1), u(k-1)] \\ y(k) &= h[x(k)] \end{aligned} \quad (11)$$

where polynomials p_1 and p_2 represent the identified polynomial ARMA model for .the conversion (y_1) and the molecular weight (y_2), respectively.

The state estimator can be obtained from eq (11) by using EKF and by introducing the disturbance state variable in order to obtain unbiased estimates as follows:

$$\begin{aligned} \begin{bmatrix} \hat{x}(k|k-1) \\ \hat{x}^w(k|k-1) \end{bmatrix} &= \begin{bmatrix} F(\hat{x}(k-1|k-1), u(k-1)) \\ \hat{x}^w(k-1|k-1) \end{bmatrix} \\ \hat{y}(k|k) &= h(\hat{x}(k|k-1) + \hat{x}^w(k|k-1)) \end{aligned} \quad (12)$$

$$\begin{bmatrix}\hat{x}(k|k)\\ \hat{x}^w(k|k)\end{bmatrix}=\begin{bmatrix}\hat{x}(k|k-1)\\ \hat{x}^w(k|k-1)\end{bmatrix}+L(k)(y_m(k)-\hat{y}(k|k)) \quad (13)$$

where $y_m(k)$ and $L(k)$ represent the output measurement and estimator gain, respectively. The output prediction equation is derived from the above state estimator with several assumptions as follows:

$$\begin{bmatrix}\hat{y}(k+1|k)\\ \hat{y}(k+2|k)\\ \vdots\\ \hat{y}(k+p|k)\end{bmatrix}=\begin{bmatrix}I\\ I\\ \vdots\\ I\end{bmatrix}\cdot\left(h(\hat{x}(k|k)-H_k\hat{x}(k|k)\right)$$

$$+\begin{bmatrix}H_kF_1(\hat{x}(k|k),u(k-1))\\ H_kF_2(\hat{x}(k|k),u(k-1))\\ \vdots\\ H_kF_p(\hat{x}(k|k),u(k-1))\end{bmatrix} \quad (14)$$

$$+\begin{bmatrix}H_kB_k^u & \cdots & \cdots & 0\\ H_k(B_k^u+A_kB_k^u) & H_kB_k^u & \cdots & 0\\ \vdots & & & \vdots\\ \sum_{i=0}^{p-1}H_kA_k^iB_k^u & \cdots & \cdots & 0\end{bmatrix}\begin{bmatrix}\Delta u(k)\\ \vdots\\ \vdots\\ \Delta u(k+p-1)\end{bmatrix}$$

We considered the following constraints in the simulation study.

Jacket inlet temperature (u_1): $55^\circ C \le T_{jin} \le 85^\circ C$

Feed flow rate (u_2): $5ml/min \le q_f \le 30ml/min$

Conversion (y_1): $0 \le X \le 0.5$

Molecular weight (y_2): $10000 \le M_w \le 60000$

$$|\Delta T_{jin}| \le 5^\circ C \quad , \quad |\Delta q_f| \le 1ml/min$$

Figure 2 shows the regulatory performance of the nonlinear model predictive controller based on the identified ARMA model (NLMPC) listed in Table 4. In this case, we assumed that the monomer concentration is increased from 4.34 mol/l to 6.51 mol/l, whereas the solvent concentration is decreased from 4.7 mol/l to 2.35 mol/l between 200 min and 300 min. In this figure, solid and dashed lines represent the case for the nonlinear ARMA and linear ARMA, respectively. As the monomer concentration is increased, both the conversion and the molecular weight are increased, however the NLMPC regulates rapidly both the conversion and the molecular weight. The model predictive controller based on the linear ARMA model (LMPC) shows the similar results as those of the NLMPC, however, for the control of the molecular weight, the NLMPC is superior to the LMPC.

The servo performance of the NLMPC is presented in Figure 3. For the control of the conversion, the LMPC shows the more rapid response than the NLMPC, however LMPC shows also oscillatory response. Since the molecular weight has the more nonlinerities than the conversion, as shown in Figure 2 and 3, the NLMPC shows the better performance for the control of the molecular weight. In the simulation study, the prediction horizon and the control horizon were 30 and 3, respectively.

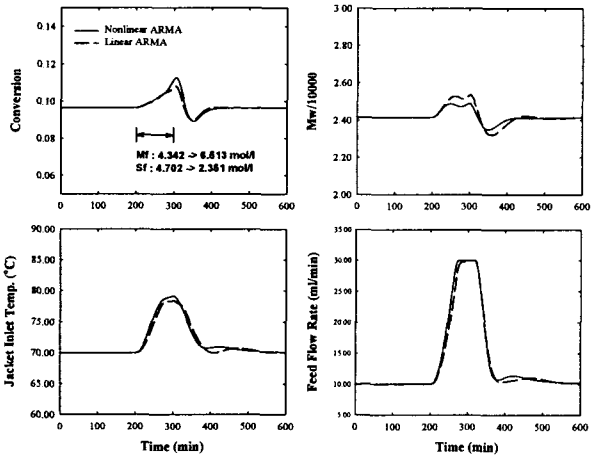

Figure 2 The regulatory performance of the NLMPC and the LMPC.

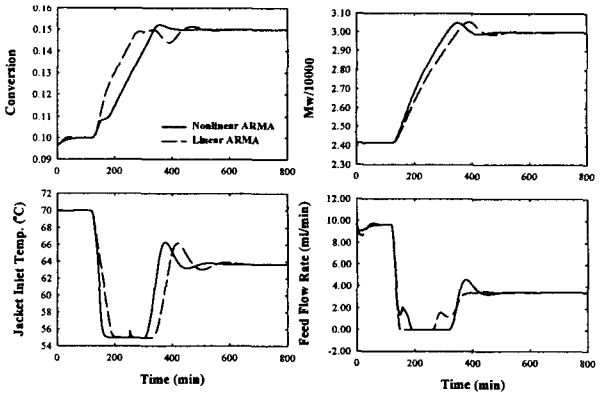

Figure 3 The servo performance of the NLMPC and the LMPC.

4. Conclusions

The multivariable nonlinear model predictive controller based on the polynomial ARMA model is developed and implemented by simulation. A polynomial ARMA model can effectively describe the dynamic behavior of polymerization system by using the stepwise model building algorithm implemented in this work. Despite the complex and nonlinear features of the polymerization reaction system, the proposed NLMPC is found to perform satisfactorily for the quality control of the continuous styrene polymerization reactor.

Acknowledgement

The authors wish to acknowledge the financial support of the Korea Research Foundation made in the program year of 1997.

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