Polypropylene Degradation Control during Reactive Extrusion

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ABSTRACT: In this work, a proportional-integral-derivative (PID) control scheme with two different tuning methods to control the degree of degradation of polypropylene (PP) during reactive extrusion is proposed. The concentration of dicumyl peroxide is taken as the manipulated variable. The molten viscosity of PP under processing is taken as the controlled variable. The degree of degradation is determined by a viscosity function derived by an off-line identification. A first-order-plus-time-delay empirical model is identified to simulate the system plant. Both Ziegler–Nichols tuned PID and internal model control (IMC)-based PID controllers are implemented on the system. Better performances in settling time and precision can be achieved using the IMC-based PID controller. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 95: 280–289, 2005

Key words: reactive extrusion; degradation; polypropylene

INTRODUCTION

Reactive extrusion involves reaction kinetics and flow dynamics and it is a highly integrated production method, the end product of which is finished in one run.[1,2] It was proposed[3–5] that introducing various peroxides at diverse controlled rates during the polypropylene (PP) reactive extrusion process can carefully control the extent of degradation. Several experimental and theoretical investigations on PP degradation control have also been studied.[6–8] In a PP degradation control process, the control variable is commonly suggested to be the melt viscosity, since the reduction of MW would relatively decrease the melt viscosity.[9–11]To track real-time changes in MW for determining the extent of degradation, it is strongly suggested to control the melt viscosity in line.[12–16] Several studies also proposed the in-line control system models of PP degradation during reactive extrusion.[16–18] Pabedinskas et al.[19] further worked on controller design and performance analysis. However, the dynamic of the reactive extrusion plant is a rather complex process, and any process identification method used in previous works can only provide a similar solution that leads to various control schemes with modified strategies implemented to perform the quality control.

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Since the process identification method (empirical modeling) is not as straightforward, a tuning method that can provide a design trade-off between control performance and robustness to model uncertainties is needed. proportional-integral-derivative (PID) controller is mostly used during polymer processing, and there are several PID controller tuning methods available. Among them, IMC claims to provide robust control.The Ziegler–Nichols tuned method should produce tuning parameters that will obtain quarter wave decay.[20–22] In this work, an attempt to assess the performances using these two tuning methods on PID controller during reactive extrusion to get a better performance is proposed. The degradation behaviors have also been shown on a series of MWD plots using a technique known as gel permeation chromatography (GPC) for verifyication.

EXPERIMENTAL

Experimental apparatus

The PP degradation control experiments were done on a single screw extruder (as shown in Fig. 1). The specifications of the single screw extruder are shown in Table I. Three ASAHI, Model TTJ-N67A, pressure transducers were used combined with temperature sensors located at the solid, molten, and the melt transition sections of the barrel, respectively, to monitor the operating conditions during extrusion. An in-line viscometer[23] was mounted between the barrel and the die to real-time estimate melt viscosity.

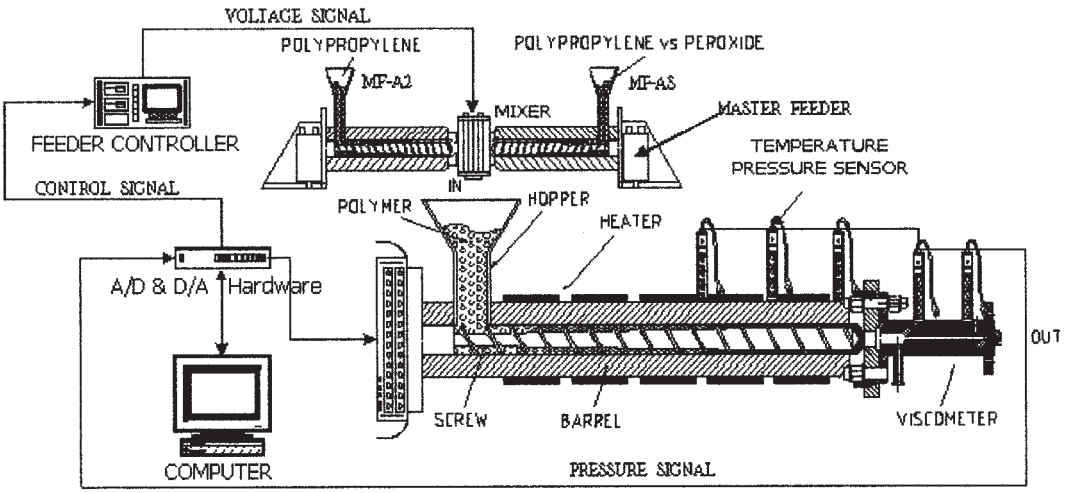

Figure 1 PP degradation control system.

The PP resin was added at controlled rates between 1.15 and 11 kg/h to the mixer via a screw feeder (Yann Bang Electrical Machinery Co., MF-A2). On the other side, the 0.2% dicumyl peroxide (DCUP)/PP premixed compound was added at controlled rates between 0.2 and 2.0 kg/h to the mixer via a MF-AS screw feeder. Then the mixture was fed into the extruder feeder through the mixer. A personal computer combined with analog-to-digital (A/D) and digital-to-analog (D/A) converters (Axiom Model AX5411) and RS232 interface was used for monitoring and controlling all of the process information including temperature, pressure, feeder controller, and screw speed.

Experimental materials

The PP, PC366–3, made by Taiwan Polypropylene Co. with specifications listed in Table II, was used as the raw material to produce different grades of PP (PC366–5, 6H31, 6331). We simplified the three grades as A, B, and C, respectively, for convenience.

DCUP, whose chemical structural formula is $[C_6H_5C(CH_3)_2]_2O_2$, with 39 ~41°C melting point made by Aldrich Chemical was used as the initiator.

CONTROL SYSTEM CONFIGURATION

The whole control system was implemented with two main steps, as shown in Figure 2, including an off-line process that derives the desired viscosity of desired grade PP and an in-line control process that takes the derived viscosity as the set point of the control system. The further procedures on the viscosity function development, process model identification, and controller design are illustrated as follows.

TABLE I
Specifications of the Single Screw Extruder

Specifications	Value
Screw diameter (mm)	45
L/D ratio	25
Compression ratio	3.37
Production output rate (kg/h)	4–35
Screw speed (rpm)	0–100

Viscosity function development

A series of flow tests of different grades at three different temperatures (see Fig. 3) were tested, in advance, to create the viscosity function. Equation (1) was identified by a numerical analysis method called Exponential Decay First-Order approximation. Table III shows parameters of the equation under different conditions.

$$\eta = Y_0 + A_1 e^{-\left(\frac{\dot{\gamma} - X_0}{t_1}\right)}, \tag{1}$$

where η denotes viscosity and $\dot{\gamma}$ is shear rate.

Process model identification

The dynamic model between viscosity and initiator concentration was empirically identified into a single-input–single-output model. The detailed procedure is divided into three steps. The first step includes evaluating the fit validity of a theoretical step response to actual system response data of a step input. So, a step change of the initiator concentration from 0.01 to 0.02 wt % at 200°C temperature set point was added in the process. The subsequent output viscosity measured in response to these changes is shown in Figure 4. Next, a first-order plus dead time model, as shown in Eq. (2), was picked to represent the process dynamic according to the step response shown in Figure 4.

TABLE II
Specifications of Different Grades of PP Made by Taiwan Polypropylene Co. Ltd.

	Unit	Test method or standard	PC366-3	PC366-5	6H31	6331
MFI	g/10 min	ASTM D1238	3	5	8	14
Density	g/cm^3	ASTM D792	0.901	0.903	0.902	0.904
Tensile stress at yield	kg/cm^2	ASTM D638	350	355	355	355
Tensile strain at yield	%	ASTM D638	10	9.0	9.0	8.8
Flexural modulus	kg/cm^2	ASTM D790	16,900	17,300	16,500	17,300
Shrinkage	%	ASTM D955	1.60	1.46	1.55	1.51
Number average MW	(g/mol)	GPC	57,551	40,015	12,532	10,304
Weight average MW	(g/mol)	GPC	688,309	208,118	138,453	126,630
Z average MW	(g/mol)	GPC	781,814	449,850	284,312	253,305

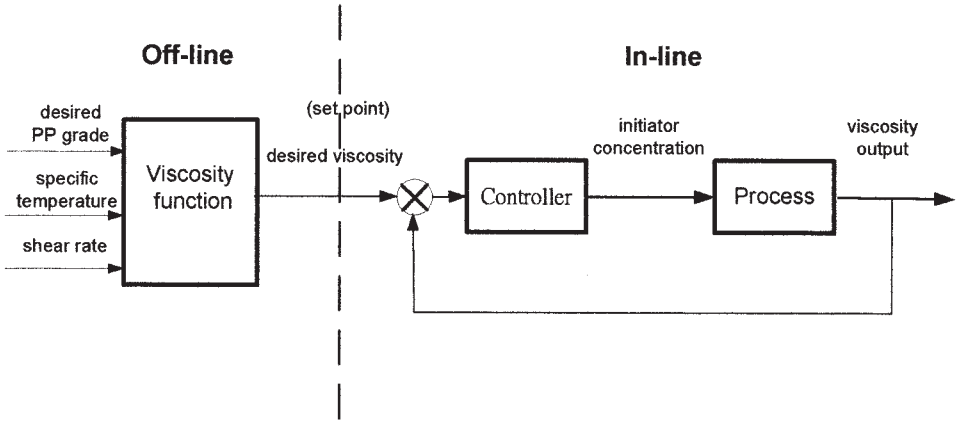

Figure 2 Control system configuration.

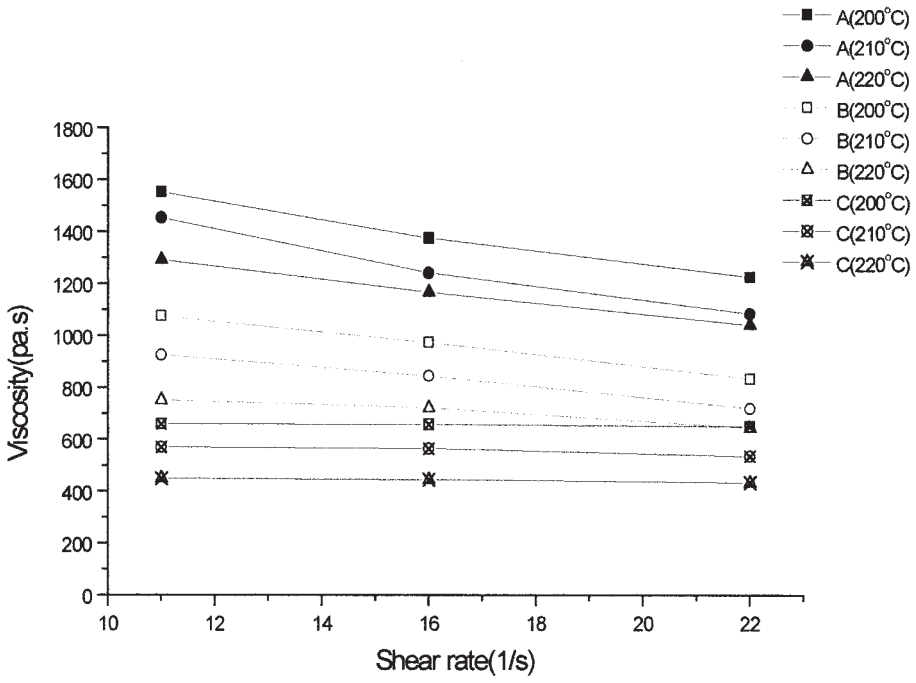

Figure 3 Shear rate versus viscosity plots of different grades of PP.

TABLE III
Parameters in Eq. (1) under Different Conditions

	Y_0	A_1	X_0	t_1
Grade A				
200 °C	909.26	644.74	11	15.48
210 °C	863.42	591.58	11	11.20
220°C	442.30	850.70	11	31.46
Grade B				
200°C	−36,604.75	37,683.42	11	1716.5
210 °C	−39,215.7	40,146.46	11	2147.7
220 °C	−5978.94	6737.71	11	693
Grade C				
200°C	−16.33	677.44	11	813.29
210°C	−1855.53	2429.09	11	797.56
220°C	−612.47	1063.63	11	723.77

TABLE IV
Parameters in Process Model

Temperature (°C)	Input changes (wt %)	K_P (Pa · s/wt %)	τ_P (s)	t_d (s)
200	0.00–0.01	−51,900	417	600
	0.01–0.02	−48,600	341	680
	0.02–0.03	−32,000	341	660
	0.03–0.04	−26,300	354	660
210	0.00–0.01	−49,200	405	600
	0.01–0.02	−48,100	367	680
	0.02–0.03	−28,000	342	640
	0.03–0.04	−27,000	380	640
220	0.00–0.01	−58,400	442	640
	0.01–0.02	−37,300	392	660
	0.02–0.03	−30,000	442	640
	0.03–0.04	−25,600	379	600

$$y(s) = \frac{K_p e^{-t_d s}}{\tau_p S + 1} u(s), \quad (2)$$

where K_p is the process gain, τ_pp is the process time constant, t_d represents the time delay, s stands for the Laplace factor, and $u(s)$ and $y(s)$ denote the process input and output, respectively. The process gain was determined based on the steady-state viscosity change over the dioxide concentration change. The time delay and time constant were derived by applying a linear best-fit regression to the data set. Table IV shows the derived parameters at three different temperature conditions. The final step in process identification is to check the fitness of the empirical model to the process step output. Figure 5 shows one comparison of the empirical model and the process step output, in which the step change of the input is from 0.01% to 0.02% wt at 210°C.

Controller design

Two different controllers include Ziegler–Nichols-tuned PID and internal model control (IMC)-based PID controllers were used to derive a better performance in the degradation control process.

Ziegler–Nichols approximation tuning rules

The Ziegler–Nichols tuning rule was proposed by Ziegler and Nichols in 1942 and provides a rule for the proportional gain, integral time, and derivative time.

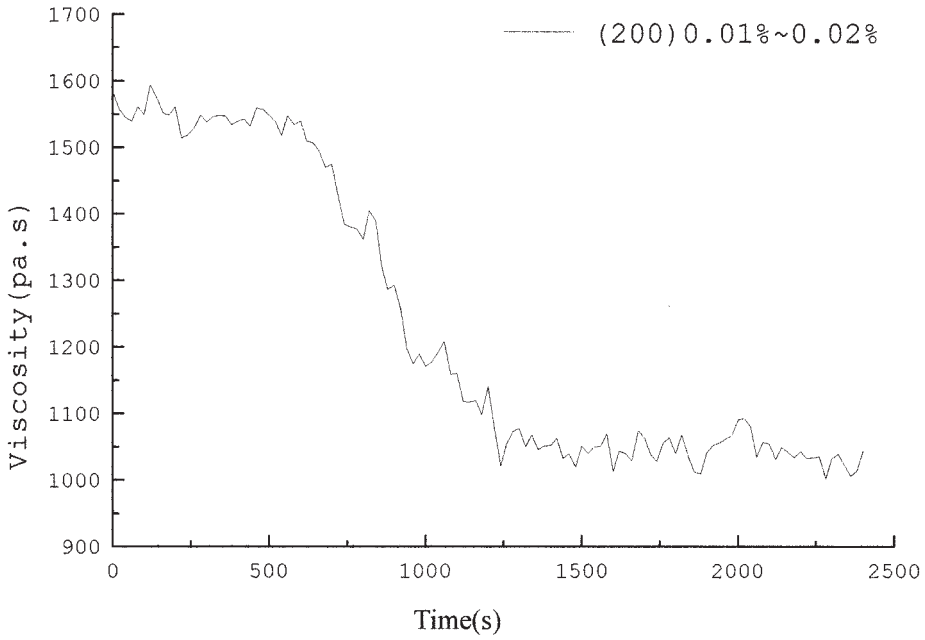

Figure 4 Step response.

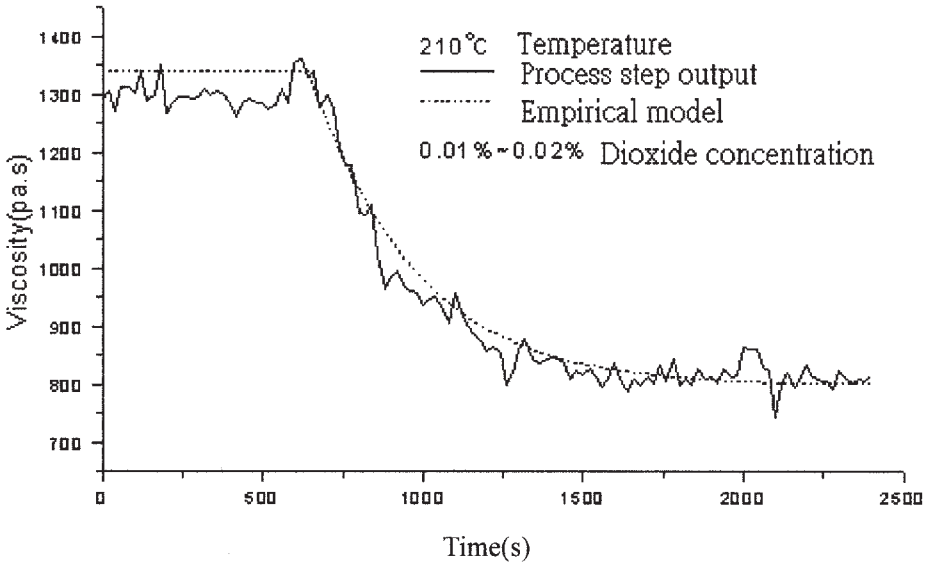

Figure 5 Comparison of the empirical model and the process step output.

TABLE V
Parameters in Ziegler–Nichols Approximate Model Tuning

Controller type	K_C	$K_I = K_C/\tau_1$	$K_D = K_C\,\tau_D$
P	−0.0000154	—	—
PI	−0.0000138	−0.0000000064	—
PID	−0.0000185	−0.000000014	−0.006

The general form of Ziegler–Nichols-tuned PID controller can be expressed as

$$G_C = K_C\left(1 + \frac{1}{\tau_I S} + \tau_D S\right). \quad (3)$$

Table V shows the parameters tuned using the Ziegler–Nichols method, where the process gain used was −38,500 (pa.s/wt %), and the delay time and time constant were chosen by values of 650 and 385 s, respectively, according to Table IV. The simulated results according to Table V at 200°C of temperature set point and 60 s of sampling time and 23 rpm of screw speed can be seen in Figure 6. The PI controller with Ziegler–Nichols tuning rule provides better performances in both overshoot and steady-state error than the others. Figure 7 shows the comparison of the results between the simulation and practical process that use the Ziegler–Nichols-tuned PI controller. The

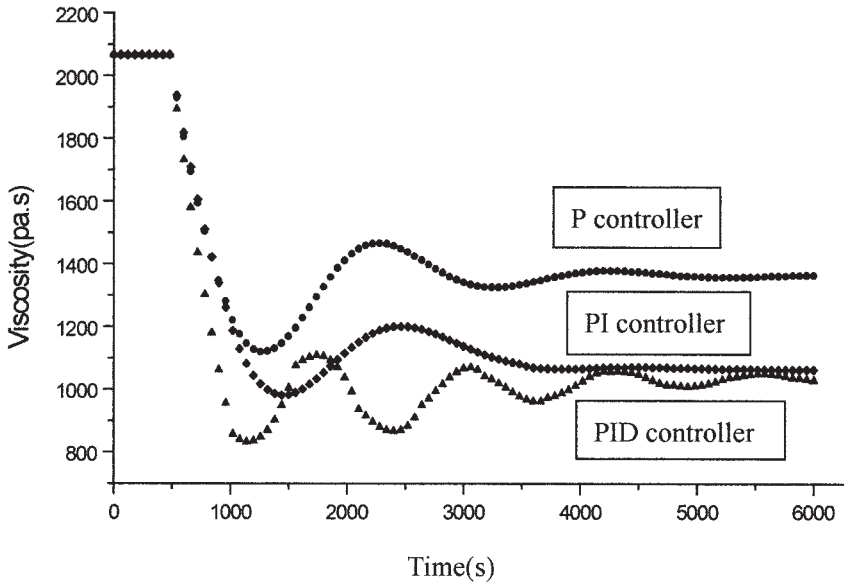

Figure 6 Simulated outputs using Ziegler–Nichols tuning P, PI, and PID controllers separately.

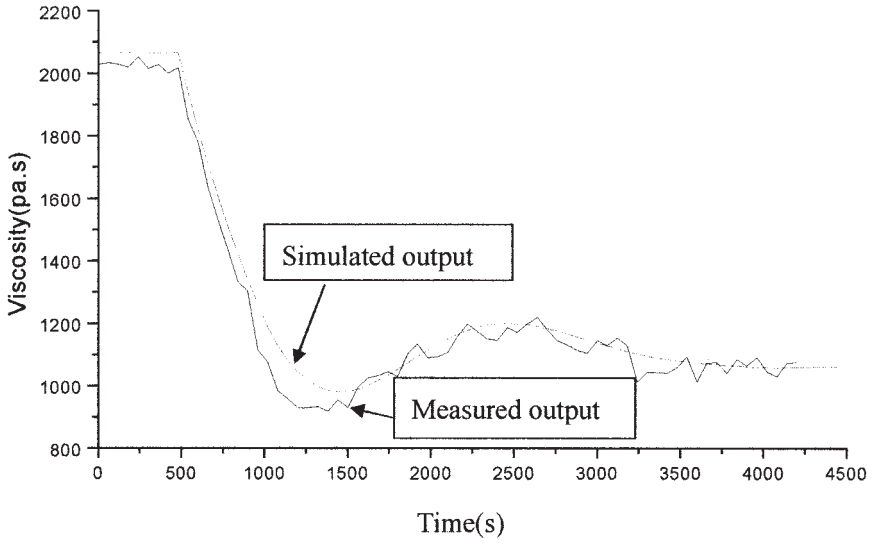

Figure 7 Comparison of simulated and measured output of the PP degradation control process with Ziegler–Nichols tuning PI controller.

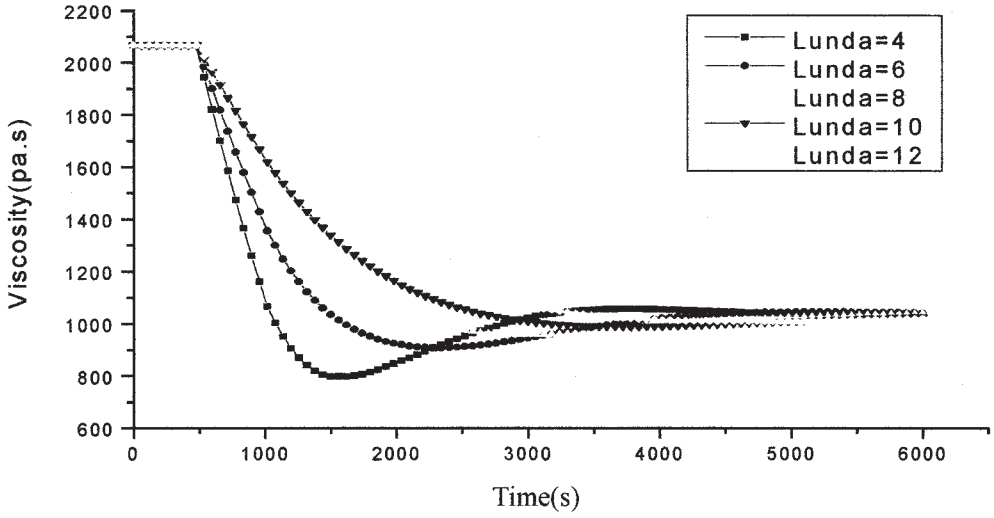

Figure 8 Simulated results of using diverse tuning parameters.

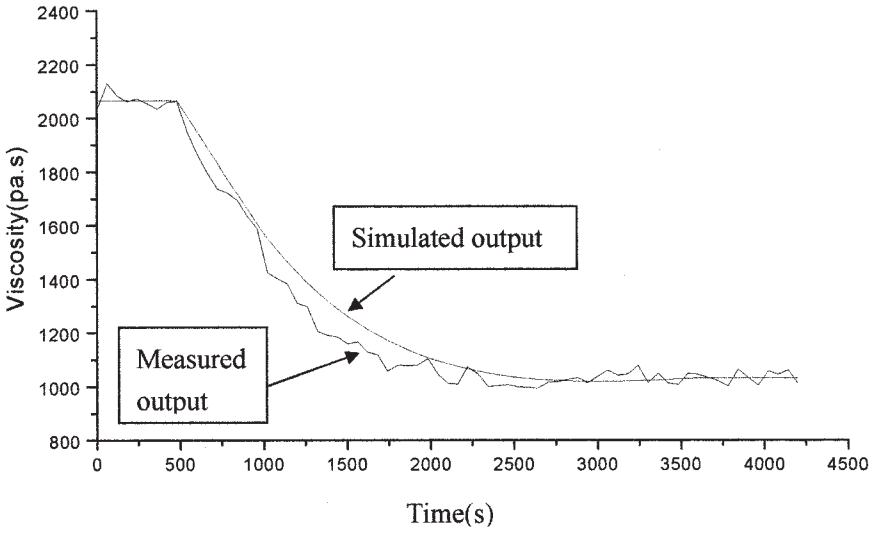

Figure 9 Comparison of simulated and measured outputs of applying the IMC-based PID controller, in which the system desired outputs are grade B.

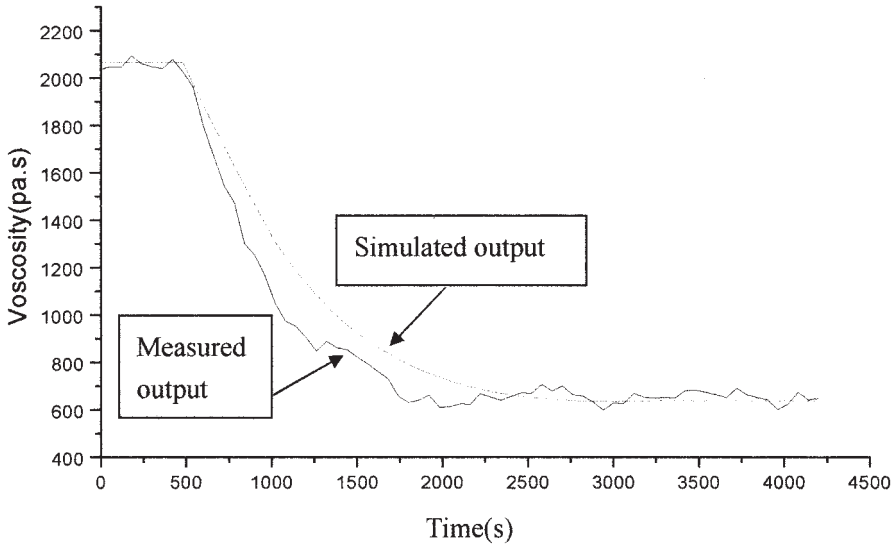

Figure 10 Comparison of simulated and measured outputs of applying the IMC-based PID controller, in which the system desired outputs are grade C.

temperature was set at 200°C and the system output was set at 1046 (pa.s), which was measured to be grade B, in the experiment.

IMC-based PID controller

The IMC-based PID controller was proposed by Rivera et al.[24] and can provide a much easier framework for the design of the robust control system. To proceed with this model, we replaced the time delay in the process model with Pade approximation. The simplified controller model would be expressed as

$$G_c = K_c\left(1 + \tau_D S + \frac{1}{\tau_1 S}\right)\left(\frac{1}{\tau_1 S + 1}\right), \tag{4}$$

where

$$K_c = \frac{2\tau_p + t_d}{2K_p(\lambda + t_d)},\ \tau_1 = \tau_p + (t_d/2),$$

$$\tau_D = \frac{\tau_p t_d}{2\tau_p + t_d} \quad \text{and} \quad \tau_1 = \frac{\lambda t_d}{2(\lambda + t_d)}.$$

To find a better performance, several values including 4, 6, 8, 10, and 12 were chosen for the tuning parameter λ.

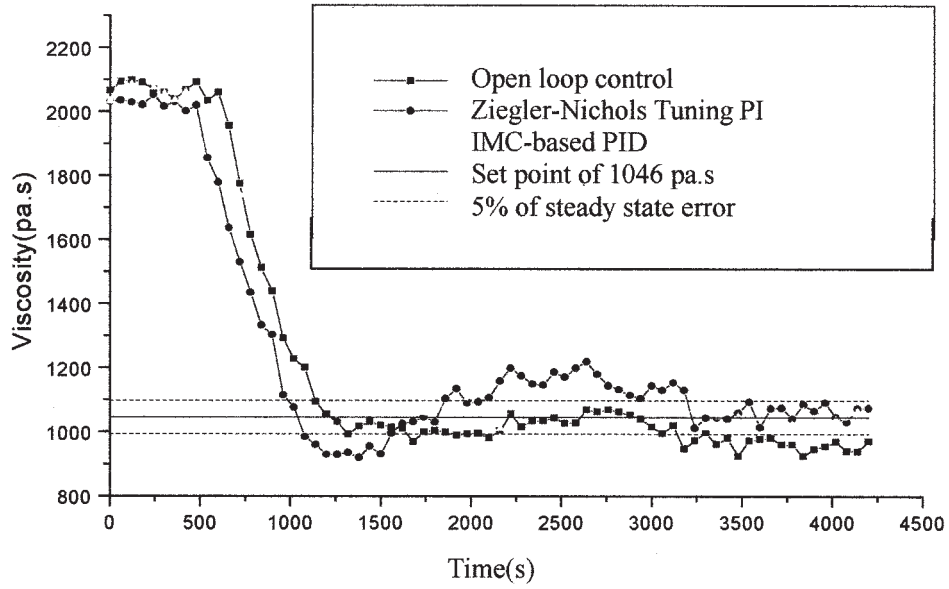

Figure 11 System responses with grade B as the system output.

TABLE VI
System Performances with Grade B as the System Output

	Open loop	Ziegler–Nichols(PI)	IMC based on PID
Maximum overshoot	—	12%	—
Rise time (s)	1260	1080	2040
Peak time (s)	—	1380	—
Settling time (s)	—	3240	2040
Standard deviation at steady state (pa · s)	46.5	11.1	13.6

Figure 8 shows the simulated results of using diverse tuning parameters, where delay time t_d = 650 s., sampling time Δt = 60 s., process gain K_p = -38,500 Pa.s wt %, and time constant τ_p= 385 s according to the system identification. We chose λ = 8 as the optimal tuning parameter for actual process control on account of its shorter settling time. Figures 9 and 10 show the results of applying IMC-based PID controller on the degradation control process with grades B and C as the system output, respectively, in which the simulated results are also presented for comparison. Figure 11 shows the measured results of applying two controllers mentioned above together with same processing conditions and set grade B as the system output. In addition, an open loop control with no controller was applying to the degradation control system too, in which the operating temperature of these results was set at 200°C and the viscosity of grade B was referred to as 1046 pa.s.

The system performances of using three different control schemes are shown in Table VI shows. As can be seen, the one using the Ziegler–Nichols (PI) control scheme possesses the fastest rise time of 1080 s, but, obtains a poor settling time of 3240 s. As for the IMC-based PID control scheme, a poor rise time of 2040 s is achieved, but a better settling time of 2040 s is also achieved. In this case, the steady-state error of both proposed control schemes is less than 2%, which is within the range of 5% that we originally designed. It is suggested that the result using the IMC-based PID controller would achieve a better performance than the others.

The case with grade A as the system output presented the same trends (see Fig. 12) under the same operating condition as mentioned above. The detailed performances are also shown in Table VII. We also used the GPC to perform MWD tests of the PP grade controlled using the IMC-based PID controller (see Figs. 13 and 14), in which the solid line in the figures refers to the MWD plot of raw material PC366–5 that we used for comparison. Detailed data of MW and MWD are shown in Table VIII.

CONCLUSION

The PP degradation can be real-time controlled to a specific grade by means of controlling its correspond-

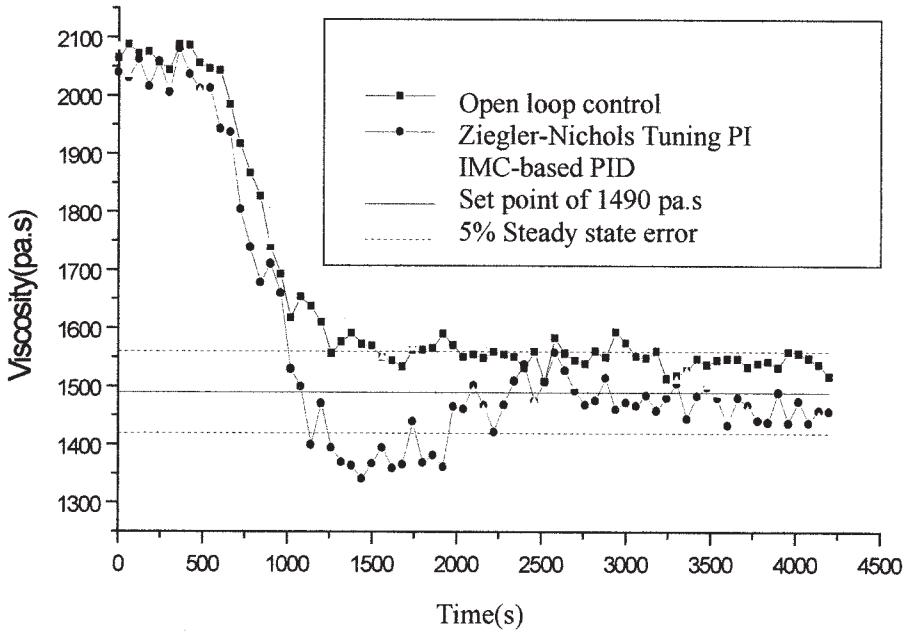

Figure 12 System responses with grade A as the system output.

TABLE VII
System Performances with Grade A as the System Output

	Open loop	Ziegler–Nichols(PI)	IMC based on PID
Maximum overshoot	—	10%	—
Rise time (s)	1260	1140	1920
Peak time (s)	—	1440	—
Settling time (s)	—	2760	1920
Standard deviation at steady state (pa · s)	62	21	9.2

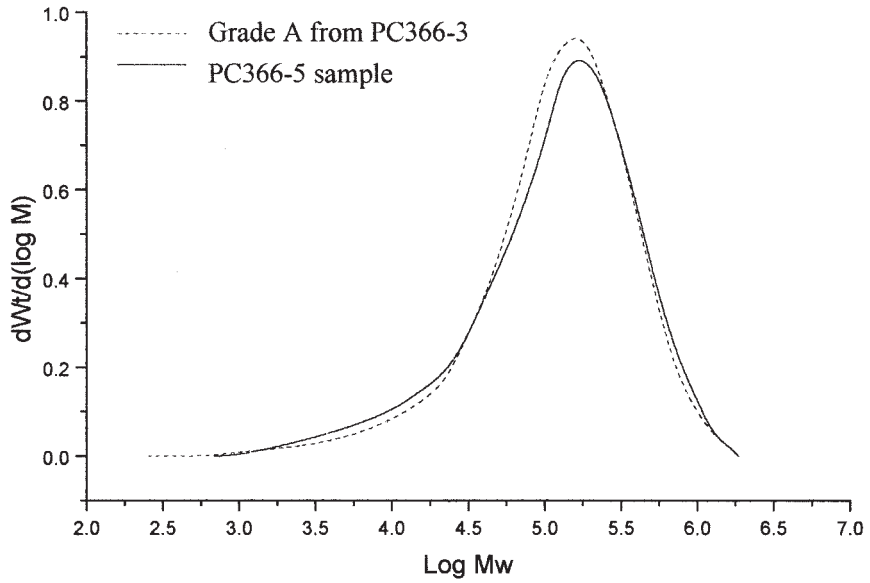

Figure 13 MWD plot of grade A controlled from PC366–3.

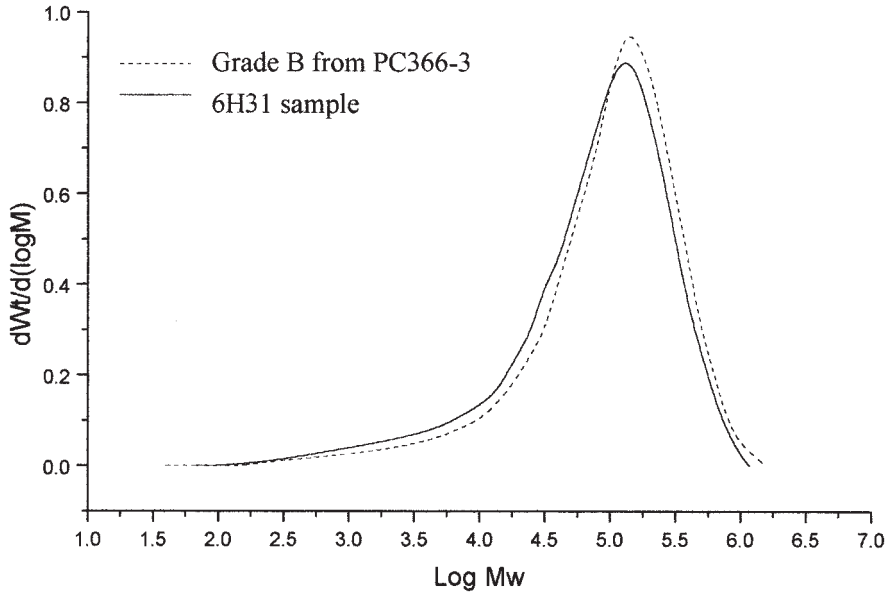

Figure 14 MWD plot of grade B controlled from PC366–3.

TABLE VIII
MW and MWD Data

	Number average MW	Weight average MW	Polydispersity
PC366_5 sample	40,015 (g/mol)	208,118 (g/mol)	5.20
Grade A from PC366-3	38,909 (g/mol)	195,171 (g/mol)	5.02
Accuracy	97.2 (%)	93.8 (%)	96.5 (%)
6H31 sample	12,532 (g/mol)	138,453 (g/mol)	11.0
Grade B from PC366-3	13,565 (g/mol)	166,618 (g/mol)	12.2
Accuracy	91.7 (%)	80 (%)	89 (%)

ing melt viscosity during the reactive extrusion process. We also announce that the specific grade of PP can be achieved by using both Ziegler–Nichols-tuned PI controller and IMC-based PID controller. The result using the IMC-based PID controller would achieve a better performance.

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