Process Optimization of Sweet Potato Pulp-Based Biodegradable Plastics Using Response Surface Methodology

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ABSTRACT: Biodegradable plastics were produced from sweet potato pulp (SPP) and cationic starch (CS) or chitosan composite (CC) by compression molding and their mechanical properties were tested. A universal testing machine, Rockwell hardness tester, and Izod impact tester were used for testing the mechanical properties (flexural strength, Rockwell hardness, and Izod strength) of the plastics. A central composite second-order design was used to study the effects of temperature, time, and moisture content on the flexural strength, Rockwell hardness, and Izod strength of SPP/CS and SPP/CC blended plastics. The flexural strength, Rockwell hardness, and Izod strength of SPP-based plastics was 101.1–305.9 kg/cm^2, R29.0–R96.7, and 0.6–3.0 kg cm cm^{-2}, respectively. Regression analysis predicted the optimal mechanical properties (flexural strength, Rockwell hardness, and Izod strength) to be attained with a 150–160°C temperature, 15–20-min reaction time, and 20–23% moisture content. © 2002 John Wiley & Sons, Inc. J Appl Polym Sci 83: 423–434, 2002

Key words: sweet potato pulp; flexural; Rockwell hardness; Izod; response surface methodology

INTRODUCTION

The most widely used polymeric materials developed in the past 50–60 years are durable and inert in the presence of microbes, thus leading to long-term performance. However, in view of the current emphasis on environmental pollution problems and in conjunction with the land shortage problems for solid waste management and pending legislation, the need for environmentally degradable polymers has arisen.[1,2]

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Table I Real Value of Coded Level of Independent Variables for Experimental Design in SPP/CS and SPP/CC

	X_i Independent Variables	X_1 Temperature (°C)	X_2 Time (min)	X_3 MC (%)
Coded value of SPP/CS	−2	130	10	10
	−1	140	15	15
	0	150	20	20
	1	160	25	25
	2	170	30	30
Coded value of SPP/CC	−2	130	10	15
	−1	140	15	20
	0	150	20	25
	1	160	25	30
	2	170	30	35

Biodegradable plastics have been developed mainly on the basis of starch,[3–8] bacteria-produced materials,[9] and natural polymers such as cellulose,[10–12] soy protein,[13–15] and zein.[16] The biodegradable materials are intended mainly for the manufacture of extruded and molded articles such as utensils and containers. Glenn and Hsu[7] processed starch in various ways to make products with some properties similar to petroleum-based plastics. Van Soest et al.[4,8] investigated the mechanical properties of maize starch-based plastics made by compression molding. Domingo and Morris[3] studied the mechanical performance of extruded cornstarch-based biodegradable plastics. Bacteria-produced polyhydroxybutyrate (PHB) and PHB hydroxyvalerate (HV) copolymers exhibit mechanical properties that equal or even exceed those of traditional thermoplastics. However, the current high price of such materials limits their use to a few exclusive applications such as in the field of biomedicine and chiral synthesis.[9] Several researchers have studied the

Table II Experimental Design Trial Compositions and Results

	Coded Variables			SPP/CS			SPP/CC		
Design Point	X_1	X_2	X_3	Flexural Strength (kg/cm^2)	Rockwell Hardness	Izod Strength (kg cm cm^{-2})	Flexural Strength (kg/cm^2)	Rockwell Hardness	Izod Strength (kg cm cm^{-2})
1	−1	−1	−1	254.37	R59.39	2.18	235.92	R55.51	1.86
2	−1	−1	1	268.02	R61.08	2.16	146.46	R51.69	1.48
3	−1	1	−1	207.28	R62.18	1.45	176.00	R77.27	2.60
4	−1	1	1	286.81	R40.41	1.40	168.12	R60.84	2.84
5	1	−1	−1	145.55	R59.46	2.26	176.26	R52.47	1.78
6	1	−1	1	308.71	R70.59	1.59	202.60	R40.46	1.38
7	1	1	−1	197.50	R74.78	1.50	222.87	R68.43	1.57
8	1	1	1	241.87	R87.15	1.76	223.90	R58.28	1.13
9	0	0	0	287.61	R66.21	2.36	246.09	R54.16	1.42
10	−2	0	0	279.65	R48.86	1.61	183.18	R31.20	1.98
11	2	0	0	230.10	R81.43	1.72	232.30	R33.34	1.35
12	0	−2	0	302.19	R38.48	1.85	210.12	R52.40	3.14
13	0	2	0	269.17	R77.25	1.60	151.81	R34.84	1.52
14	0	0	−2	170.60	R66.02	1.82	156.71	R58.32	3.05
15	0	0	2	194.39	R38.85	1.57	121.64	R28.30	1.38

Table III Regression Coefficients of Approximate Polynomials for Response Variables in Experimental Design

	SPP/CS			SPP/CC		
Coefficient	Flexural Strength	Rockwell Hardness	Izod Strength	Flexural Strength	Rockwell Hardness	Izod Strength
B_0	−596.619	327.865	−33.135	−868.232	−1783.779	7.388
B_1	12.142	−1.708	0.470	17.068	22.032	−0.064
B_2	−2.802	−11.251	−0.219	−21.792	7.725	0.490
B_3	6.338	−8.542	0.278	−0.571	11.775	−0.264
B_{11}	−0.066	−0.007	−0.002	−0.096	−0.074	0.001
B_{12}	0.033	0.124	0.002	0.265	0.007	−0.006
B_{22}	0.042	−0.101	−0.006	−0.654	−0.177	0.009
B_{13}	0.286	0.109	−0.001	0.312	−0.005	−0.002
B_{23}	−0.265	−0.111	0.005	0.281	−0.054	0.003
B_{33}	−0.989	−0.155	−0.006	−1.071	−0.230	0.008
R^2	0.7432	0.8176	0.7812	0.8609	0.4908	0.6719

properties of blends of biomaterials and synthetic polymers.[17,18] In principle, some of the properties of starch can be significantly improved by blending it with synthetic polymers. However, most of the synthetic polymers are immiscible with starch at the molecular level.[13]

Sweet potato pulp (SPP) is a byproduct of sweet potato starch (SPS) extraction processing and is economically feasible. SPS is the major source in preparing Tangmyun (starch noodle), which is a popular food in Korea, China, and Japan. Solid waste from the SPS plant has caused serious environmental problems and has a high treatment cost in these countries. One method reported to reduce greatly the load of solid waste on landfills is the development of biodegradable plastics.[3] SPP consists mainly of cellulose (95%) and minor components, such as ash (3%), protein (0.4%), and fat (0.6%). Cellulose suspended in water becomes anionic at the surface from the actions of hydroxyl groups and a trace amount of carboxyl groups. On the other hand, chitosan is insoluble in water, and its salts, such as those with hydrochloric acid or acetic acid, become water-soluble and cationic. Chitosan has good affinity for several anionic biopolymers, such as cellulose, due to reactions between anionic groups in cellulose and cationic groups in chitosan.[19] Cationic starch (CS) was prepared by a reaction with aminating agents in the presence of alkali, and the resulting amine group has a cationic charge.[20] In previous research, CS has been used to increase the strength of papers.[21,22] These materials are known to be sensitive to processing conditions such temperature, pressure, time, and water content.[15] Three parameters, i.e., temperature, time, and moisture content (MC), greatly affected the mechanical properties of SPP/CS and SPP/chitosan composite (CC) blended plastics in our previous study.[23]

Response surface methodology (RSM) is a useful statistical technique for building empirical models, which use sequential experimental techniques to survey a domain of interest and to focus on the most important variables and their effects. Most of the RSM applications come from areas such as chemical or engineering processes, industrial research, and biological investigations, with emphasis on optimizing a process or system. The main advantage of RSM is the reduced number of experimental runs needed to provide sufficient information for statistically acceptable results.[24–31]

The objectives of this study were to produce SPP/CS and SPP/CC blended plastics, to determine their mechanical properties (flexural strength, Rockwell hardness, and Izod strength), and to optimize the process condition by response surface methodology.

EXPERIMENTAL

Materials

The following materials were used to prepare the SPP/CS and SPP/CC blended plastics: SPP (dried,

Table IV Mechanical Properties (Flexural Strength, Rockwell Hardness, and Izod Strength) of Biodegradable Plastics

Plastics	Thickness (mm)	Mechanical Properties		
		Flexural Strength (kg/cm^2)	Rockwell Hardness	Izod Strength ($kg\ cm\ cm^{-2}$)
Biodegradable plastics in this study				
SPP/CS	5.5–7	115.5–305.9	R31.1–R96.7	1.1–2.4
SPP/CC	5.5–7	101.1–251.0	R29.0–R71.5	0.6–3.0
Composite plastics from the literature				
Nylon/carbon yarn[a]	10.1	680–4840	—	—
LDPE/rice starch[b]	2.4	64–192	—	—
LDPE/potato starch[b]	2.4	28–189	—	—
Palm leaves/STMP pulp[c]	3.4	49.3	—	—
Soy protein isolate[d]	3.6–4	—	R118.4	—
HDPE[e]	3.6–4	—	R40	—
PP[f]	3.25	—	—	1.5–1.7
PPS/EGMA[g]	—	—	—	1.2–3.3
PVDF[h]	—	—	R110–115	—
PE[h]	—	560	—	9.0
PET[h]	—	2000	—	11.0
PS[h]	—	1200	—	10.7
PC[h]	—	1300	—	11

[a] Kuo and Fang (2000)[32]; data at 220–240°C for 15 min.
[b] Arvanitoyannis et al. (1998)[33]; data at 110–120°C for 15 min.
[c] Mansour et al. (1998)[34]; data at 160°C for 10 min.
[d] Schilling (1995)[15]; data at 140°C for 6 min.
[e] Data were reported by Shah (1984).[35]
[f] Data were reported by Tai et al. (2000).[36]
[g] PPS/EGMA, poly(phenylenesulfide)/poly(ethylene-*stat*-glycidyl methacrylate) Lee and Chun (1998)[37]; data at 315–320°C by injection moder.
[h] *Plastics Technology Handbook* (1987).[36]

ground, and screened with a 100-mesh sieve; Kwakji Agricultural Ind. Co., Cheju Island, Korea); cationic starch (degree of substitution with amine group of 0.04; Samyang Genex Co., Ltd., Inchon, Korea); chitosan (origin, red crab; degree of deacetylation >85%; MW 130,000; Biotech Co., Inc., Mokpo, Korea); *K*-carrageenan (Myungshin Hwasung Co., Ltd., Yangsan, Kyungnam, Korea); and glutaraldehyde and lactic acid (Showa Co., Ltd., Tokyo, Japan).

Preparation of SPP/CS and SPP/CC

SPP and CS were mixed in the ratio 90:10 (w/w) for 20 min using a mixer. CS was added in a paste. Chitosan was dissolved in a 2.5% aquous lactic acid solution with a homogenizer at room temperature. After SPP (94.5%), *K*-carrageenan (2.5%), and the chitosan solution (2.5%) were blended together, glutaraldehyde (0.5%) was added to the mixture.[23] The water content of the mixture was determined by a moisture determination balance (Kett Engineering Co., Ltd., Korea) by measuring the loss in weight after heating at 105°C for 30 min.

Preparation of the Test Specimens

The required quantity of the mixture (270 g) was applied to a mold (220 × 250 × 60 mm). The mold cavity was filled with the materials and covered with the top plate and placed between the platens of the press. On the surface of the mold, a pressure of 200 kg/cm^2 was applied. The mold was heated at 130–170°C for 10–30 min. The mold was then cooled to room temperature at 10–15°C for 15min and the mold content was released. The specimens were equilibrated for 2 days at 20°C and 50% relative humidity. The dimensions of the test samples were as outlined in the American

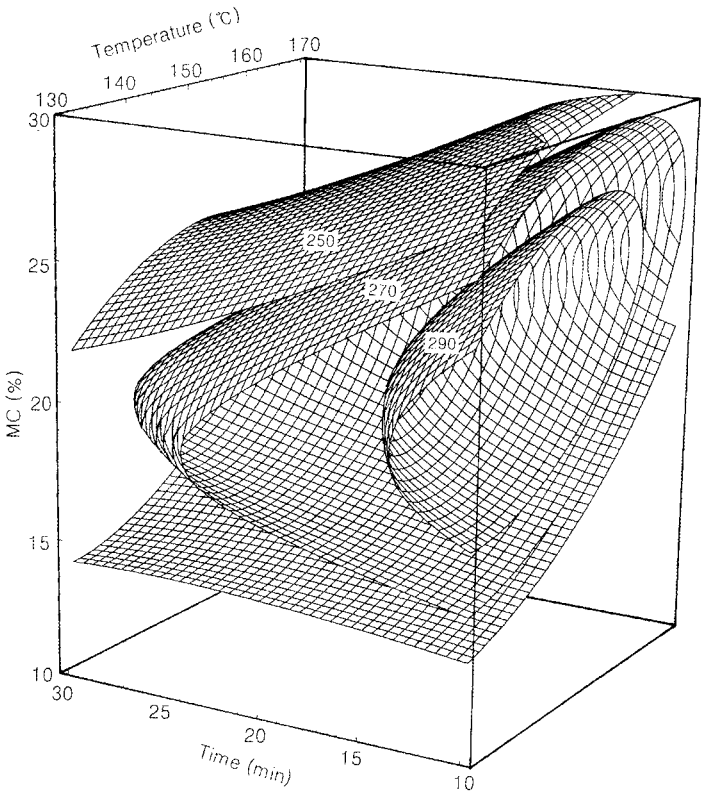

Figure 1 Computer-generated contour surface of flexural strength of SPP/CS blended plastic at 250, 270, and 290 kg/cm^2.

Society for Testing Materials (ASTM) standard method.[39–41]

Test Methods

Flexural Test

The flexural test was performed according to ASTM D 790 with an SFM-20 universal testing machine.[39] The specimen was supported on two rollers applied symmetrically near the ends and loaded from above by one roller in the middle. The span length was 90 mm and the span-to-thickness ratio was 15. The crosshead speed was 5 mm/min. The results were calculated from the data obtained from five parallel tests.

Rockwell Hardness Test

Rockwell hardness was measured using the ASTM D 785 standard method. Rockwell hardness can differentiate the relative hardness of different types of a given plastic. The procedures entailed use of a 0.5-in.-diameter, stainless-steel spherical indenter (Rockwell hardness tester DTR-300).[40] First, a minor load of 10 kg was applied to each sample; the Rockwell R hardness was subsequently measured after 15 s of continuous application of a 60-kg major load. Rockwell hardness measurements were obtained on five specimens of each formulation. Each specimen was indented five times.

Izod Impact Test

The toughness of the materials was characterized by Izod impact tests on unnotched samples due to their brittleness. The Izod test was performed according to ASTM D 256 with a universal impact tester with a hammer (2 J) at room temperature.[41] The span length was 65 mm and span-to-thickness ratio was 10. The results were calculated from the data obtained from five parallel tests.

Experimental Design

To assess the effects of operating parameters on the mechanical properties of SPP/CS and SPP/CC blended plastics, a central composite rotatable response surface experimental design was used.

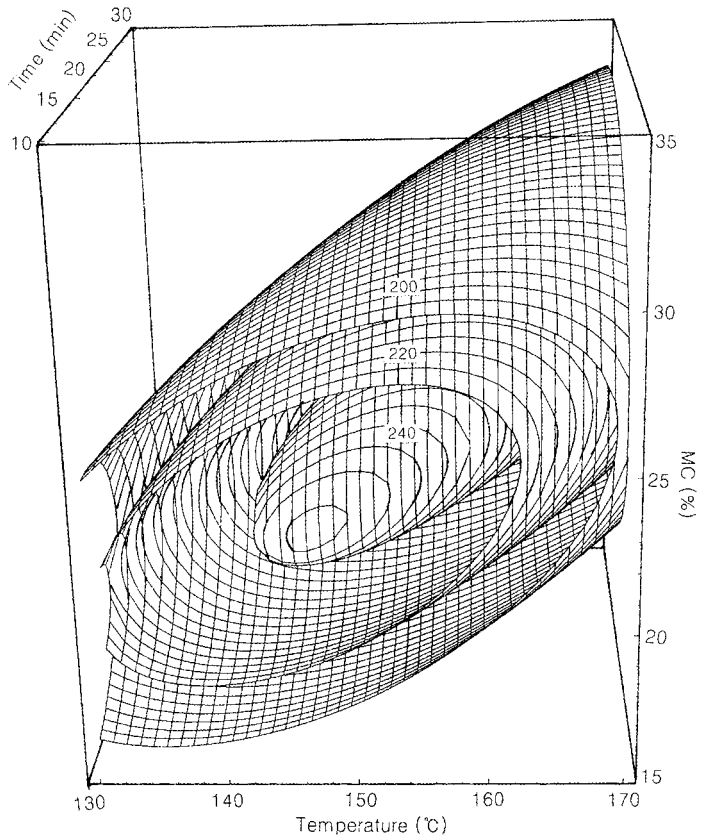

Figure 2 Computer-generated contour surface of flexural strength of SPP/CC blended plastic at 200, 220, and 240 kg/cm^2.

The experimental design was a modification of central composite design for three variables at five levels each. Temperature (X_1), time (X_2), and MC (X_3) were independent variable factors. The coded values of the independent variables were −2, −1, 0, 1, and 2. The actual values and the corresponding coded values of three factors are given in Table I. The ranges and center-point values of the three independent variables were chosen based on preliminary trials. Among all possible combinations, 15 conditions were carried out in a random order. The flexural strength, Rockwell hardness, and Izod strength of plastics were dependent variable responses. The value of each dependent response was the mean of five replications. Data were analyzed to fit the following second-order equation to all dependent Y variables: $Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{11} X_1^2 + \beta_{12} X_1X_2 + \beta_{22} X_2^2 + \beta_{13} X_1X_3 + \beta_{23} X_2X_3 + \beta_{33} X_3^2$. Coding of the natural variables, temperature, time, and MC, at the center point were taken to be 150, 20, and 20 (in SPP/CS) and 25 (in SPP/CC), respectively. The response surface regression (RSREG) procedure of the Statistical Analysis System (SAS, 1996) program[42] was used to fit quadratic polymonial equations to experimental data and to test the models for goodness of fit.

RESULTS AND DISCUSSION

Mechanical Properties

The mechanical properties of SPP/CS and SPP/CC blended plastics performed by following the experimental design are summarized in Table II. The regression coefficients (B_i) are presented in Table III. The mechanical strengths (flexural strength, Rockwell hardness, and Izod strength) of the SPP-based plastics are shown in Table IV and compared with other composite plastics. The mechanical strengths of the SPP-based plastics were lower than those of commercial plastics such as PE and PET but close to those of biopolymer composite plastics such as LDPE/rice starch and LDPE/potato starch composite plastic.[33] The flex-

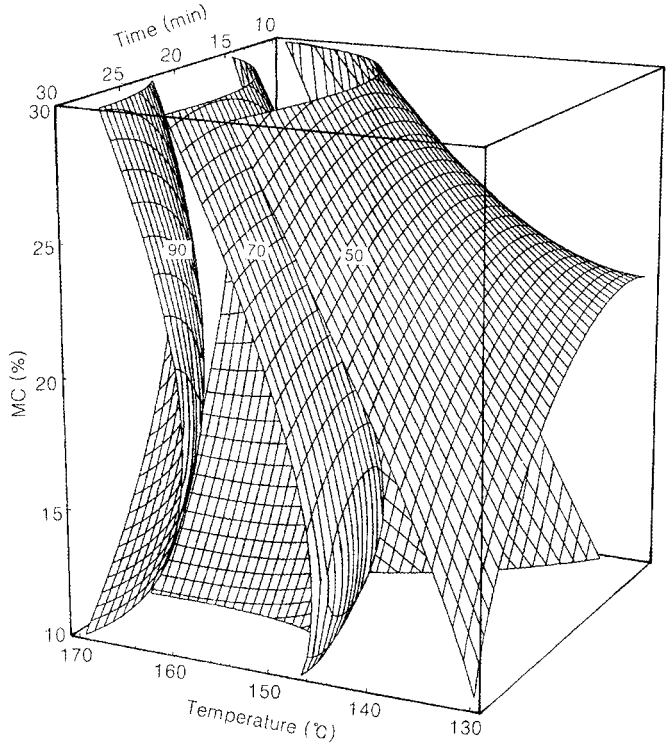

Figure 3 Computer-generated contour surface of Rockwell hardness of SPP/CS blended plastic at R50, R70, and R90.

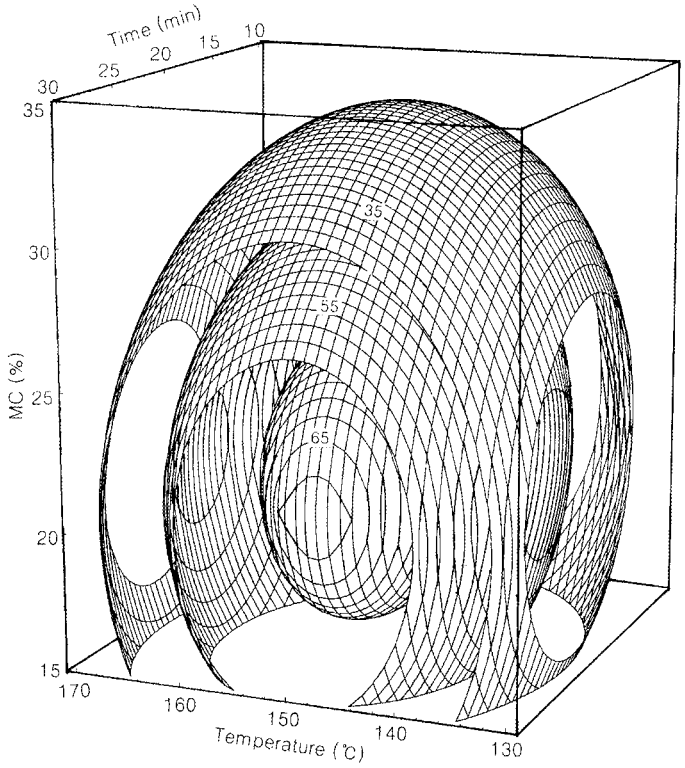

Figure 4 Computer-generated contour surface of Rockwell hardness of SPP/CC blended plastic at R35, R55, and R65.

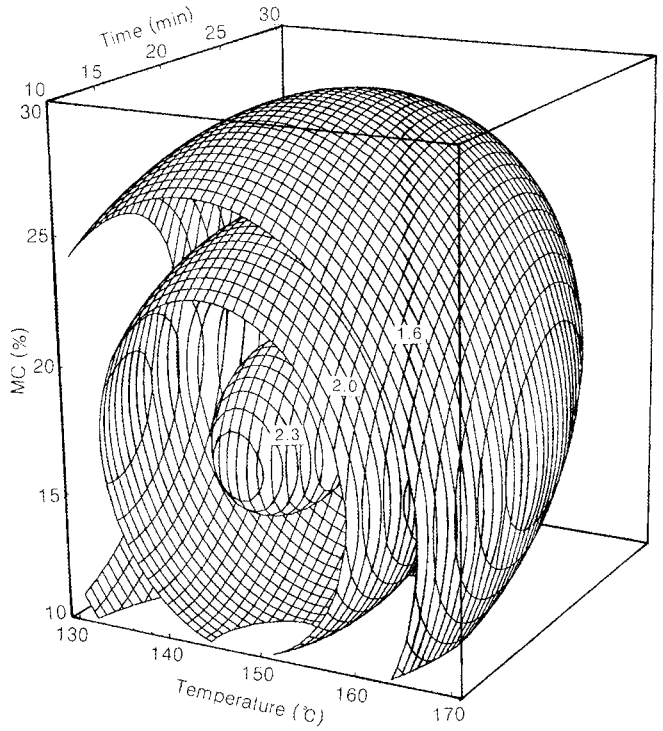

Figure 5 Computer-generated contour surface of Izod strength of SPP/CS blended plastic at 1.6, 2.0, and 2.3 kg cm cm^{-2}.

ural strength of the SPP-based plastics was 101.1–305.9 kg/cm^2, which is lower than those of PE, PP, PET, and nylon/carbon yarn composite plastic. But the flexural strength of the plastic was higher than those of LDPE/rice starch (64–192 kg/cm^2) and LDPE/potato starch (28–189 kg/cm^2) composite plastics. The Rockwell hardness of SPP-based plastics was R29.0–R96.7, lower than those of polystyrene, nylon 6-6, PVDF, and soy protein isolate plastics but was higher than that of HDPE (R40). The Izod strength of SPP-based plastics was 0.6–3.0 kg cm cm^{-2} and close to those of polypropylene (1.5–1.7 kg cm cm^{-2}) and PPS/EGMA composite plastics (1.2–3.3 kg cm cm^{-2}). The predictive models were modified and used to create four-dimensional response surfaces within the experimental region using a Mathematica program (version 3.0, Wolfram Research, Inc. 1988–96). Based on the predicted models, four-dimensional surfaces for the flexural strength, Rockwell hardness, and Izod strength of SPP/CS and SPP/CC blended plastics are shown in Figures 1–6.

Flexural Strength

The flexural strength of SPP/CS blended plastics depends mostly on the MC (X_3) as its quadratic effect as well as the linear effect being significant. The other factors that also contribute to the flexural strength include the linear effect of temperature (X_1) and its interaction with MC (X_1X_3) (Table III). Based on the predicted models, the four-dimensional surfaces for the flexural strength of the SPP/CS blended plastics are shown in Figure 1. Decreasing temperature and time increased the flexural strength of the SPP/CS blended plastics at 20–25% MC (Fig. 1). The four-dimensional response surfaces for the flexural strength of SPP/CC blended plastics are shown in Figure 2. The flexural strength of the SPP/CC blended plastics was entirely lower than that of the SPP/CS blended plastics. In contrast to the SPP/CS blended plastics, the flexural strength of the SPP/CC blended plastics increased with increasing temperature. Similar observations were reported by Kuo and Fang.[32] and Paetau et al.[14] A regression model fitted for the flexural strength in SPP/CS and SPP/CC gave a reasonably good fit: $R^2 = 0.74$ and 0.86, respectively.

Rockwell Hardness

The Rockwell hardness of SPP-based plastics was lower than those of commercial plastics such as

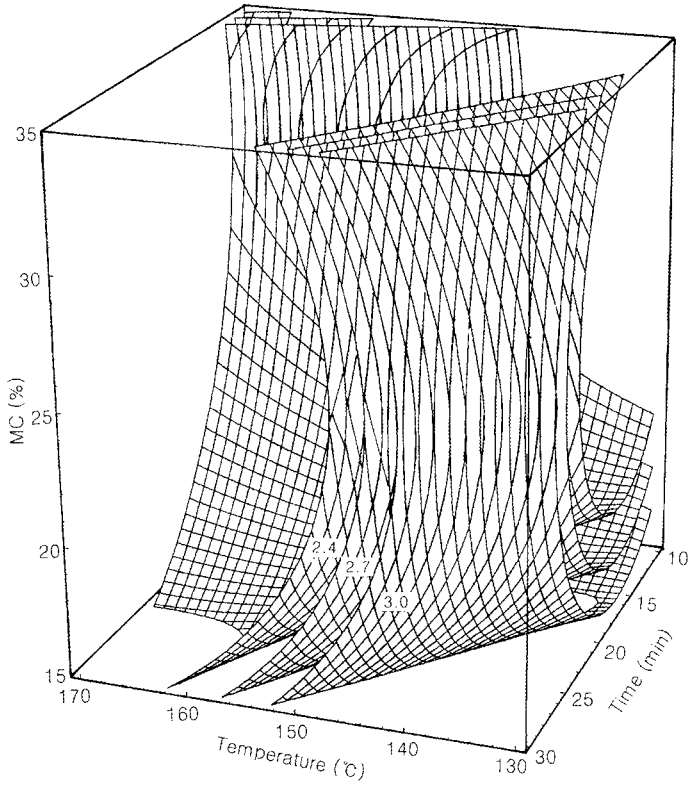

Figure 6 Computer-generated contour surface of Izod strength of SPP/CC blended plastic at 2.4, 2.7, and 3.0 kg cm cm^{-2}.

PVDF, as shown in Table IV. The Rockwell hardness of SPP/CS blended plastics was R31.1–R96.7, which was higher than those of SPP/CC blended plastics (R29.0–R71.5). The four-dimensional response surfaces for the Rockwell hardness of SPP/CS blended plastics are shown in Figure 3. The Rockwell hardness of the SPP/CS blended plastics increased with increasing temperature and reaction time. In contrast to the flexural strength, the reaction time has a significant effect on the Rockwell hardness of SPP/CS blended plastic (Table III). At high temperature (170°C), an increase in time increases the Rockwell hardness of plastic (Fig. 3). The four-dimensional response surfaces for the Rockwell hardness of the SPP/CC blending plastics are shown in Figure 4. The Rockwell hardness of the SPP/CC blended plastics increased with temperature and MC approaching close to 150°C and 20–25%, respectively. A regression model fitted for Rockwell hardness in SPP/CS gave a reasonably good fit (R^2 = 0.82). However, the R^2 of the regression equation of the Rockwell hardness in the SPP/CC blended plastic was 0.49. Probably, a power model is necessary to describe adequately the Rockwell hardness dependence on the temperature, time, and MC for SPP/CC blended plastic.

Izod Strength

The Izod strength of the SPP-based plastics was much lower than those of PE, PET, PS, and PC and was close to those of PP (1.5–1.7 kg cm cm^{-2}) and PPS/EGMA composite plastic (1.2–3.3 kg cm cm^{-2}). The four-dimensional response surfaces for the Izod strength of SPP/CS blended plastics are shown in Figure 5. The Izod strength of the SPP/CS blending plastic reached its maximum with MC and temperature approaching close to 20% and 150°C, respectively. The Izod strength of the SPP/CS blended plastic (1.1–2.4 kg cm cm^{-2}) was between that of the SPP/CC blended plastic (0.6–3.0 kg cm cm^{-2}). The flexural strength, Rockwell hardness, and Izod strength of SPP/CS was usually higher than those of SPP/CC. It may be caused by binding forces between SPP and cationic starch or chitosan. Chitosan is the copolymer of *N*-acetyl-D-glucosamine and D-glucosamine and has a linear structure. The starch consists of two major components: amylose (28%),

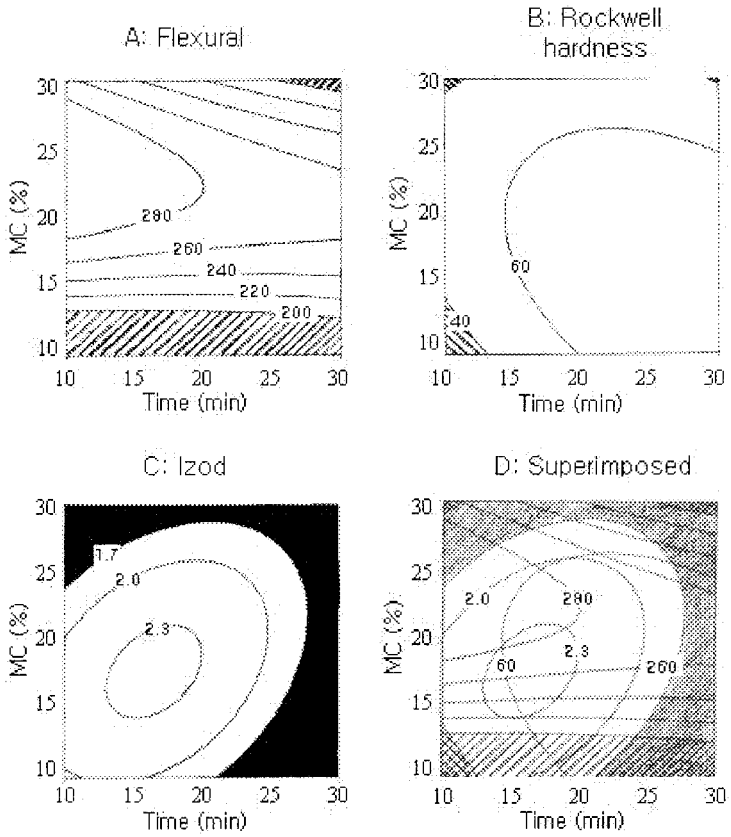

Figure 7 Superimposed contour map for overall optimization of response variables in mechanical properties of SPP/CS blended plastic.

a mostly linear alpha-D-(1-4)-glucan, and amylopectin (72%), an alpha-D-(1-4) glucan which has alpha-D-(1-6) linkages at the branch point. Therefore, SPP can bind more strongly with CS than with chitosan. A regression model fitted for the Izod strength in SPP/CS gave a reasonably good fit ($R^2 = 0.78$). However, R^2 of the regression equation of the Izod strength in the SPP/CC blended plastic was 0.67. Probably, a power model is necessary to describe adequately the Izod strength dependence on temperature, time, and MC for SPP/CC blended plastic.

Optimum Process Conditions

The predictive models of the mechanical properties (flexural strength, Rockwell hardness, and Izod strength) of these plastics were used for further examination of the system behavior and localization of the optimum conditions. The investigation was focused on finding the optimal conditions of SPP/CS and SPP/CC blended plastics, which would satisfy the mechanical properties of flexural strength $>$ 200 kg/cm^2, Rockwell hardness $>$ R40, and Izod strength $>$ 1.7 kg cm cm^{-2}. The lower limit of the 200 kg/cm^2 flexural strength, R40 Rockwell hardness, and 1.7 kg cm cm^{-2} Izod was set so that the product would be similar those of commercial LDPE/starch blended plastics, HDPE, and PP, respectively (Table IV). The limit of 200 kg/cm^2 or more of the flexural strength was set and the area with less than 200 kg/cm^2 flexural strength is shaded [Fig. 7(A) and Fig. 8(A)]. The limit of R40 or more Rockwell hardness was set, and the area with less than R40 Rockwell hardness is shaded [Fig. 7(B) and Fig. 8(B)]. Accordingly, a constraint of 1.7 kg cm cm^{-2} was set for the Izod strength, and the area with values less than this limit is also shaded [Fig. 7(C) and Fig. 8(C)]. The three plots [Fig. 7(A–C) for SPP/CS and Fig. 8(A–C) for SPP/CC] were then superimposed to produce the plot of Figure 7(D) and Figure 8(D), where the not-shaded region fulfills three requirements (flexural strength $>$ 200 kg/cm^2, Rockwell hardness $>$ R40, and Izod strength $>$ 1.7 kg cm cm^{-2}) and can be used as a guide for a satisfactory process. From the super-

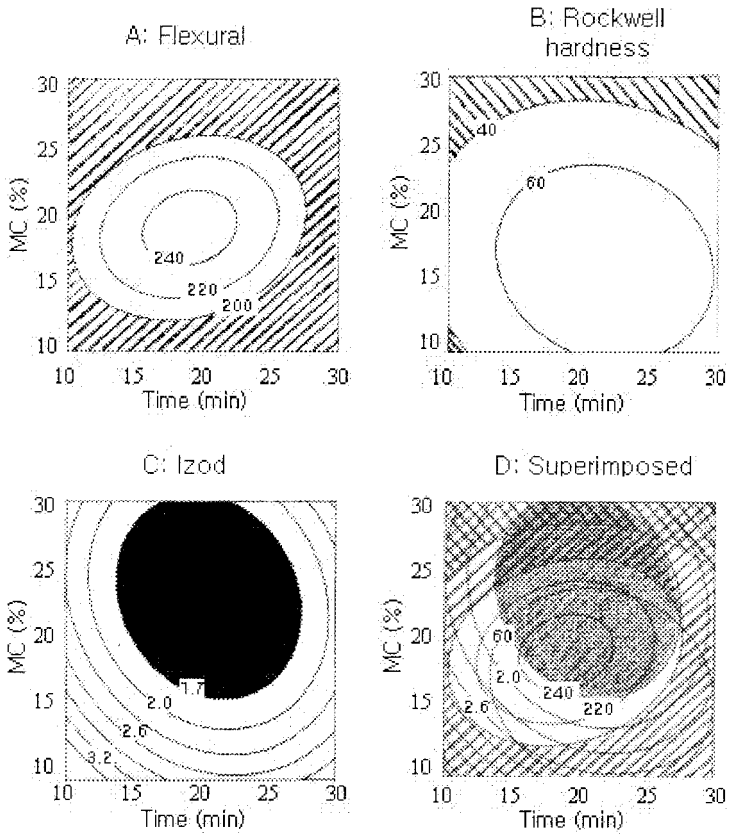

Figure 8 Superimposed contour map for overall optimization of response variables of mechanical properties of SPP/CC blended plastic.

imposed plots [Fig. 7(D) and Fig. 8(D)], it is seen that for a process under constant temperature (150°C) there are many different combinations of time and MC (not-shaded region in Fig. 7(D) and Fig. 8(D)], which would result in satisfactory mechanical properties. The optimum conditions for SPP/CS and SPP/CC blended plastics were 17–23% MC for a 16–22-min reaction time and 15–20% MC for a 13–16-min reaction time, respectively. Within this optimum range, the characteristics for SPP/CS blended plastics were a flexural strength of 260–280 kg/cm^2, Rockwell hardness of R60–R70, and Izod strength of 2.0–2.1 kg cm cm^{-2}. For SPP/CC blended plastics, the values were a flexural strength of 220–240 kg/cm^2, Rockwell hardness of R55–R60, and Izod strength of 1.8–2.0 kg cm cm^{-2}.

CONCLUSIONS

RSM was used to investigate the optimal processing condition of SPP-based plastics. Prediction models were developed for flexural strength, Rockwell hardness, and Izod strength as a function of temperature, reaction time, and MC. Processing conditions yielding an optimum process (flexural strength $>$ 200 kg/cm^2, Rockwell hardness $>$ R40, and Izod $>$ 1.7 kg cm cm^{-2}) were determined, and the local topography of the system near the predicted optimal point was developed.

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