Study on the Structure of Nylon 6 Films Iodinated Before Forming. II. Investigation of the Crystallization Behavior through Thermal Analysis

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ABSTRACT: Two kinds of amorphous nylon 6 films iodinated before forming from the powders iodinated with 0.2*N* and 1.0*N* I_2/KI aqueous solutions were prepared by a melt-press, and isothermally treated at 20 to 80°C for 1 day to 20 days. Thermal analyses were performed to investigate mainly the crystallization behavior on the treatment. The DSC thermograms for the treated films exhibit three temperature-groups of endothermic peaks at 60 ~ 70°C, 105 ~ 120°C, and higher than 150°C, which may be associated with the melting of the complex crystal, the relaxed γ-crystal, and the relaxed α-crystal, respectively. The film containing less I_2/KI and treated at the higher temperature exhibits the peaks associated with the more stable type of crystal. The peak temperature generally increases with the treating temperature and time. On the occasion of there being two peaks associated with the γ-crystal and the α-crystals, ΔH for the α-crystal increases while that for the γ-crystal decreases with increasing the treating time. The TG curves indicate two temperature-zones of weight loss by the volatilization of I_2 from I_5^- and the decompositions of I_3^- and nylon 6. With increasing treating temperature, the % weight loss by the volatilization of I_2 decreases, and consequently the temperature of the weight loss by the decomposition of nylon 6 increases. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 94: 1062–1069, 2004

Key words: nylon; films; crystallization; thermal analysis

INTRODUCTION

There are many reports on the structure of nylon 6 iodinated with aqueous solutions of I_2/KI, such as on the transition of crystal type from α to γ by iodination and deiodination,[1–5] on the crystalline structure and iodine arrangement in nylon 6-iodine complex,[6–11] and on the steps of complex formation on systematically increasing the content of I_2/KI.[12,13] From them, it is clear that polyiodide ions can penetrate into the crystalline phase as well as amorphous phase and interrupt the intermolecular hydrogen bonds and form a complex with nylon 6 chains, consequently separating the chains and enhancing chain mobility.

Several applications of the preceding effect of iodination to the drawability enhancement of nylon 6 have been attempted.[8,12] With the object of applying the iodination to the forming process as well, we have prepared a new type of nylon 6-iodine complex films, namely, the films iodinated before forming (IBF films) by melt-pressing the powders previously iodinated with I_2/KI aqueous solutions. Their structure has been discussed compared with the films iodinated after forming (IAF films) in the first part of this study.[13] Its principal conclusion was that the IBF films have practically amorphous structure, merely containing insignificant crystalline portions in a relaxed state that may have been created mainly with the iodide-ions-free segments of nylon 6 chains in the course of the film forming procedure.

We found that the IBF films, which had been very flexible just after its preparation, slowly stiffened with the lapse of time at room temperature, and suspected, through an X-ray diffractometry on the stiffened films, that the stiffening must result from the crystallization of the films. This seemed to be unlikely because we anticipated that the large quantity of polyiodide ions complexing with nylon 6 molecules in the IBF films seriously interrupt the crystallization. To investigate the slow crystallization behavior of the amorphous IBF films, therefore, we have isothermally treated the amorphous IBF films at selected temperatures for selected terms from 1 day to 20 days and performed thermal analyses on them. In this article, the results are discussed.

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EXPERIMENTAL

Materials and preparation of IBF films

The nylon 6 powder (Allied Signal, Morristown, NJ) and the iodination procedure were identical with

those described in the first part of this study.[13] To prepare two kinds of IBF films with different contents of I_2/KI, the nylon 6 powders iodinated with 0.2*N* and 1.0*N* I_2/KI aqueous solutions were used. The ultimate weight gains were 194.9% and 207.6%, respectively. The procedure and conditions of the film preparation, except the cooling rate, were identical with those described in the first part of this study.[13] The cooling rate was enhanced by an attachment for water-circulation to prevent crystallization to the utmost.

Isothermal treatments

The films were isothermally treated by immersing in silicone oil (Dow Corning Korea, Seoul, Korea) at 20 to 80°C for 1 day to 20 days. The lower and upper bounds of the treating temperature were selected in consideration of the room temperature and the highest temperature free from danger of volatilization of iodine molecules and degradation of nylon 6 molecules, respectively. After treatment, the films were rinsed with an *n*-heptane (extra pure Yakun Pure Chemicals, Osaka, Japan) to remove the silicone oil on their surfaces and air-dried.

Measurements

Differential scanning calorimetry (DSC) and thermogravimetry (TG) were performed by a TA DSC 2910 and a TA TG 2950, respectively, under the atmosphere of N_2 at heating rate of 20°C/min. In the case of DSC, stainless steel open pans were used to allow the evaporation of iodine molecules. X-ray diffraction scans were obtained by a Rigaku D/max-III-A X-ray diffractometer with Cu-Kα radiation and smoothed by an Origin 6.0 (Savitzky-Golay).

RESULTS AND DISCUSSION

Crystallization behavior

Figure 1 shows the DSC thermograms of the untreated IBF films that are very flexible even at room temperature. There is no noticeable sign on the thermograms, which shows that the films are completely amorphous. In the previous study,[13] the IBF films containing identical quantity of I_2/KI have indicated a very weak endothermic peak showing their slight crystallinity. Such an increase of the degree of amorphism in this experiment must be due to the higher cooling rate.

It has been known that amorphous nylon 6 generally indicates an endothermic glass-transition slope at around 50°C and an exothermic crystallization peak at around 170°C on its thermogram. But the films in this study do not indicate them on their thermograms: Judging from very high flexibility of the films even at room temperature, it is evident that the glass-transition temperature is lower than the lowest temperature of the test (40°C). The polyiodide ions in the films may act as a plasticizer to reduce the glass-transition temperature of the films. Chuah and colleagues[8] have reported that a DSC for a nylon 6 film iodinated with 1.0*N* I_2/KI solution showed the glass-transition at a lower temperature of 32.5°C, although the film was crystalline. And the lack of crystallization may be due to the interruption by the polyiodide ions in the films. That is, within the course of the test, the polyiodide ions complexing with the nylon 6 chains interrupt the access and arrangement of the nylon 6 chains and their segments into crystal.

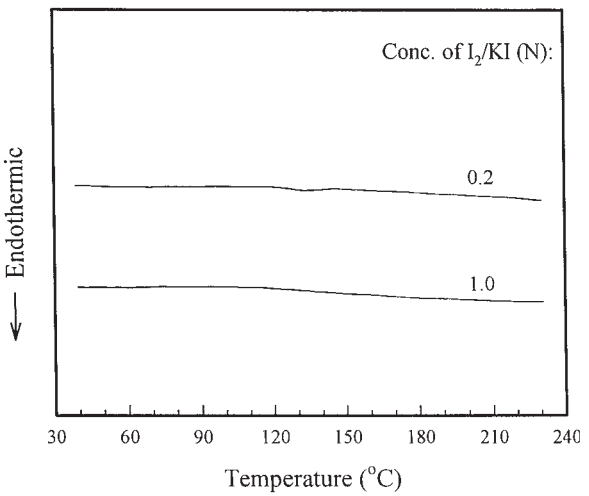

Figure 1 DSC thermograms of the untreated IBF films.

Films from the powders iodinated with 0.2*N* I_2/KI solution

The DSC thermograms of the IBF films prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution and treated at 20, 40, 60, and 80°C are shown in Figures 2, 3, 4, and 5, respectively.

Lee and colleagues[12,13] have performed the DSC analysis for nylon 6-iodine complex films prepared by immersing an α-nylon 6 film in aqueous solutions of I_2/KI with various concentrations, and reported the results as follows: The DSC thermograms indicate three kinds of melting peaks at around 65°C, 85°C, and higher than 120°C (higher than 140°C in the first part of this study[13]), which are associated with the portions of the complex crystals with I_3^- ions, the complex crystals with I_5^- ions, and the α-crystals retained in relaxed state, respectively. The thermograms for IBF films indicate only a very little endothermic peak at around 155°C, which may be associated with the melting of the relaxed α-crystals.[13] These were supported by an X-ray diffractometry and an infrared analysis.

In the Figures, there are three groups of endothermic peaks at temperature ranges of 60 ~ 70°C (in Figs.

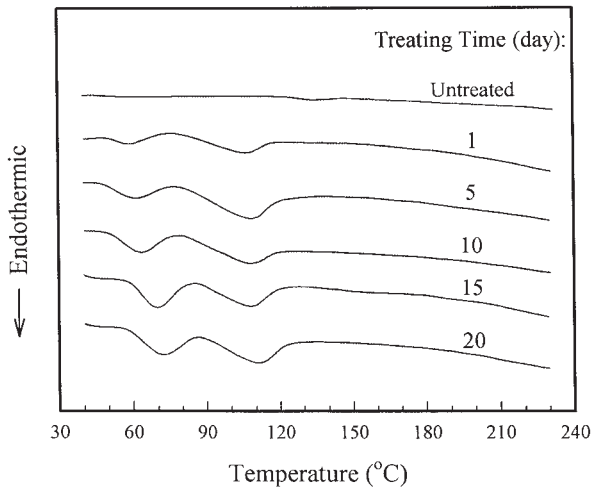

Figure 2 DSC thermograms of the IBF films, prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution, treated at 20°C for various periods of time.

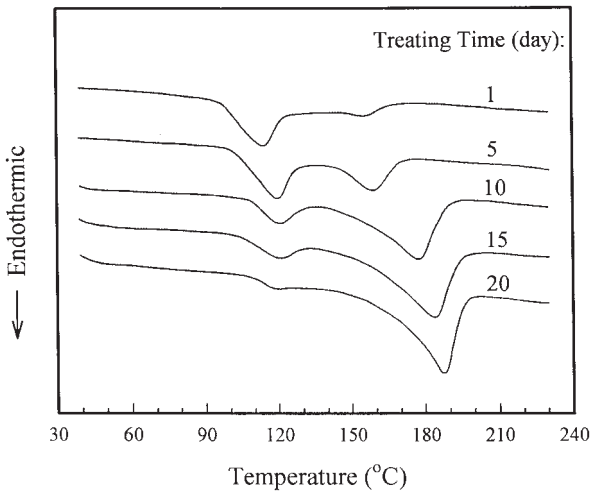

Figure 4 DSC thermograms of the IBF films, prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution, treated at 60°C for various periods of time.

2 and 3), 105 ~ 120°C (in Figs. 2, 3, and 4), and higher than 155°C (in Figs. 4 and 5). Judging by comparing the peak temperatures with those in the previous study,[13] the lower-temperature group and the higher-temperature group must be associated with the melting of the complex crystal with I_3^- and the relaxed α-crystal, respectively. But the middle-temperature group has never been observed in the previous studies.[12,13] We have presumed this new group of peaks to be associated with the melting of relaxed γ-crystals on the basis of the following details: First, it appears in the films treated at relatively low temperatures(20, 40, and 60°C) while the higher-temperature group associated with the relaxed α-crystals appears in the films treated at relatively high temperatures (60 and 80°C). Second, its peak temperature range is lower than that of the relaxed α-crystals. It is well known that the γ-crystal is formed at lower temperature and for shorter time and its melting point is lower than the α-crystal.[14]

In addition, we have tried to confirm our presumption by an experimental method as follows: A γ-crystal-rich nylon 6 film, prepared by the method of iodination and deiodination[1–5] and confirmed by an X-ray diffraction, was iodinated with an aqueous solution of 0.5*N* I_2/KI, and the DSC for it was performed under identical conditions with this experiment. Its thermogram, shown in Figure 6, indicated two recognizable endothermic peaks at around 80°C and 115°C; the lower-temperature peak must be associated with the

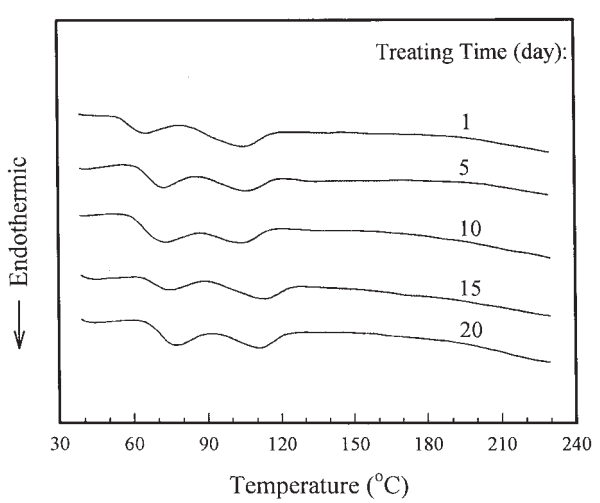

Figure 3 DSC thermograms of the IBF films, prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution, treated at 40°C for various periods of time.

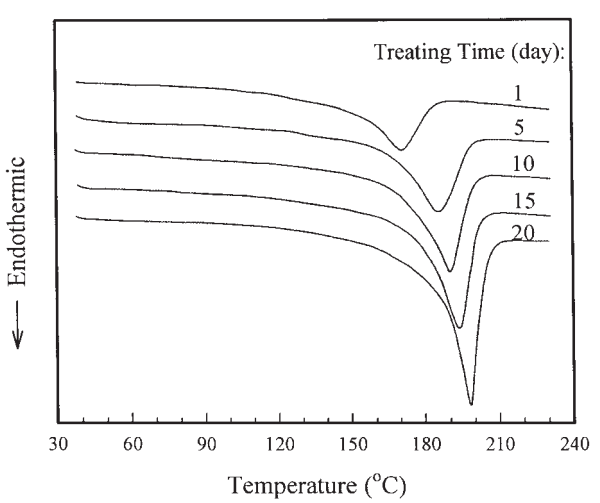

Figure 5 DSC thermograms of the IBF films, prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution, treated at 80°C for various periods of time.

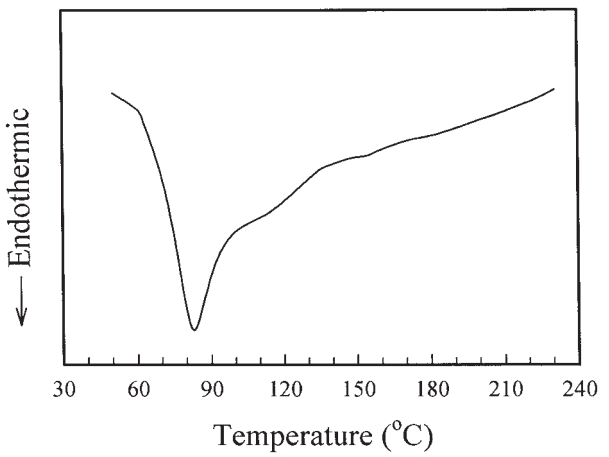

Figure 6 DSC thermogram of the γ-rich nylon 6 film iodinated with 0.5*N* I_2/KI aqueous solution.

portion of the complex crystals.[12,13] The higher-temperature peak, which is coordinate with the middle-temperature peaks in Figures 2–5, may be associated with the melting of the γ-crystals retained in relaxed state. It may be given as a conclusion that the amorphous IBF film can crystallize into three different types of crystals, that is, the nylon 6-iodine complex crystal, the relaxed γ-crystal, and/or the relaxed α-crystal, according to treating temperature.

Table I shows the maximum peak temperature (T_{max}) and energy of fusion (ΔH) obtained from each endothermic peak.

The effects of treating temperature are as follows: The films treated at 20 and 40°C, 60°C, and 80°C have the complex crystals and relaxed γ-crystals, the relaxed γ- and α-crystals, and only the relaxed α-crystals, respectively, which shows that the higher the treating temperature, the more stable the resultant crystals. In comparison among the samples treated for identical periods of time, the higher the treating temperature, the higher the T_{max}; which may result from the higher crystallization temperature that elevates the degree of crystal perfection. The dependence of ΔH on the treating temperature is not consistent in the groups of the lower-temperature peaks and the middle-temperature peaks. In the group of the higher-temperature peaks, in comparison between the samples treated for identical periods of time, ΔH for the sample treated at 60°C is obviously greater than that for the sample treated at 80°C. It is generally known that ΔH and fraction crystallinity are in proportion to each other. Therefore, a determination of the whole fraction crystallinity for each film is desirable for a significant discussion of the dependence of ΔH on the treating temperature, but the ignorance of the standard heat of fusion for each type of crystal makes it impossible. Anyway, the variation of ΔH is more consistently affected by the treating time than the treating temperature.

The effects of the treating time are as follows: With increased treating time, T_{max} increases more or less in all cases, especially in a large amount in the case of the higher-temperature peak associated with the relaxed

TABLE I
Maximum Peak Temperature (T_{max}) and Energy of Fusion (ΔH) of the Endothermic Peaks for the Films Prepared from the Powders Iodinated with 0.2N I_2/KI Solution

Treating condition		Lower-temp. peak		Middle-temp. peak		Higher-temp. peak	
Temp. (°C)	Time (day)	T_{max} (°C)	ΔH ($\times10^{-2}$J/g)	T_{max} (°C)	ΔH ($\times10^{-2}$J/g)	T_{max} (°C)	ΔH ($\times10^{-2}$J/g)
	1	59	5	106	16	—	—
	5	63	13	107	28	—	—
20	10	64	18	107	24	—	—
	15	71	24	109	22	—	—
	20	74	23	112	22	—	—
	1	66	15	105	17	—	—
	5	74	17	105	12	—	—
40	10	74	23	106	10	—	—
	15	76	10	111	12	—	—
	20	77	20	113	11	—	—
	1	—	—	114	38	155	9
	5	—	—	121	39	159	29
60	10	—	—	121	20	176	43
	15	—	—	121	18	177	56
	20	—	—	121	9	184	69
	1	—	—	—	—	170	40
	5	—	—	—	—	184	59
80	10	—	—	—	—	189	79
	15	—	—	—	—	193	96
	20		—	—	—	197	136

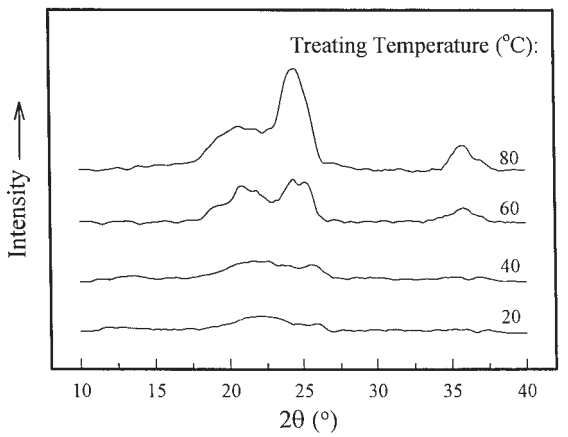

Figure 7 X-ray diffractometer scans of the IBF films, prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution, treated for 20 days at various temperatures.

α-crystals. In the cases of 20 and 40°C, the variations of ΔH for the lower-temperature and middle-temperature peaks are not consistent except a little increase at the initial time. This result suggests that a transition of the complex crystals to the γ-crystals with the lapse of time was not generated. In the case of 60°C, however, ΔH for the middle-temperature peak decreases while that for the higher-temperature peak increases; which shows that not only a creation of the α-crystals but also the transition of the γ-crystals generated mainly at the initial time to the α-crystals with the lapse of time gradually proceeded. These may result from the fact that the γ-crystal is different in its constituent elements from the complex crystal but identical with the α-crystal. That is, both the γ- and α-crystals must consist mainly of the iodide-ions-free segments of nylon 6 chains,[13] but the complex crystal must consist of the nylon 6 molecular segments complexing with abundant polyiodide ions. In the case of 80°C, ΔH for the higher-temperature peak associated with the relaxed α-crystals increases alone.

The X-ray diffractometer scans of the samples treated for 20 days are shown in Figure 7. Though the scans are not clear and the diffraction peaks are relatively weak because the structure of the films is in relaxed state by the iodination, they are helpful to support the above-mentioned discussion of the thermograms. It is well known that the nylon 6 α-crystal exhibits strong (200) and (002 + 202) diffraction peaks at $2\theta = 20.2°$ and 24.0°, respectively, and γ-crystal exhibits strong (200) and (002 + 202) diffraction peaks at $2\theta = 21.8°$ and 22.7°, respectively. Several papers[6–11] have reported that the nylon 6-iodine complex crystal exhibits diffraction peaks of its own at around $2\theta = 5.3°$, 11.5°, and 23.0°. In Figure 7, a diffraction peak at $2\theta = 25 \sim 26°$ is excluded from the discussion because it is likely due to the KI crystals ($2\theta = 25.3°$).[12] The films treated at 20°C and 40°C, which have been estimated by their thermograms (Figs. 2 and 3) to have the complex crystals and the γ-crystals, a very weak and broad plateau at $2\theta = 21°$ through 23° can fairly well support the existences of the complex crystal ($2\theta = 23°$) and the γ-crystal ($2\theta = 21.8°$ and 22.7°) in the films. The films treated at 60°C and 80°C, which have been estimated by their thermograms (Figs. 4 and 5) to have the α-crystals nearly alone, exhibit peaks at around $2\theta = 20.5°$ and 24.5°, which are regarded to be slightly shifted from the peaks at $2\theta = 20.2°$ and 24.0° being related to the α-crystal, and another peak at around $2\theta = 35 \sim 36°$, also being related to the α-crystal ($2\theta = 36°$).[15] With increased treating temperature, the intensities of the plateau and the peaks increase, which may have a close relationship to the increase of T_{max} and/or ΔH on the thermograms.

Films from the powders iodinated with 1.0*N* I_2/KI solution

The DSC thermograms for the IBF films prepared from the powders iodinated with 1.0*N* I_2/KI aqueous solution and treated at 20, 40, 60, and 80°C are shown in Figures 8, 9, 10, and 11, respectively.

The thermograms also indicate the above-mentioned three groups of endothermic peaks, but the details are somewhat different from the case of the films from the powders iodinated with 0.2*N* I_2/KI solution: There are a group of the lower-temperature peaks (the complex crystals), a group of the middle-temperature peaks (the relaxed γ-crystals), and two groups of the middle-temperature peaks (the relaxed γ-crystals) and the higher-temperature peaks (the relaxed α-crystals) on the thermograms for the films treated at 20 and 40°C, 60°C, and 80°C, respectively.

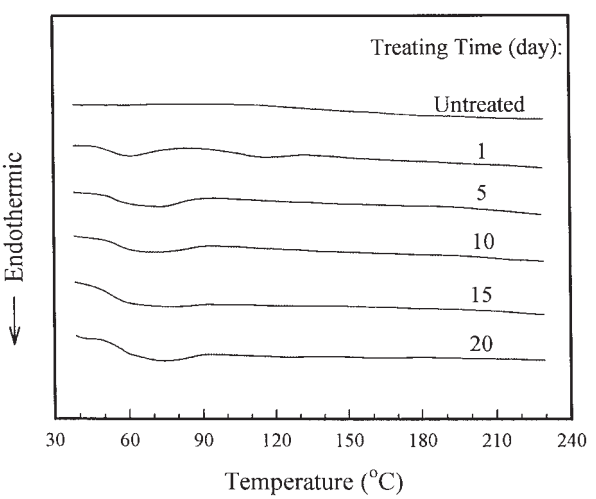

Figure 8 DSC thermograms of the IBF films, prepared from the powders iodinated with 1.0*N* I_2/KI aqueous solution, untreated and treated at 20°C for various periods of time.

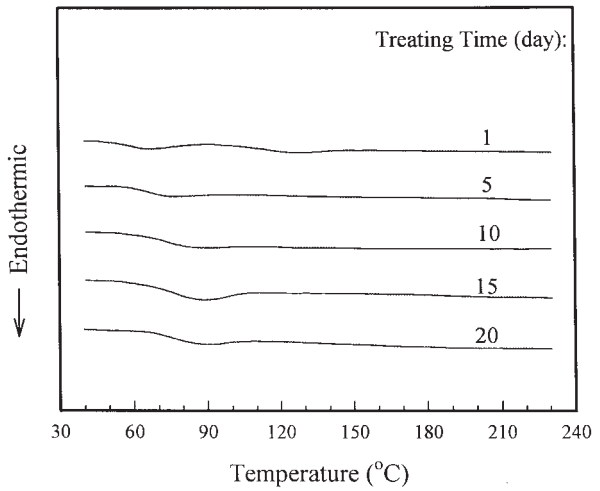

Figure 9 DSC thermograms of the IBF films, prepared from the powders iodinated with 1.0*N* I_2/KI aqueous solution, treated at 40°C for various periods of time.

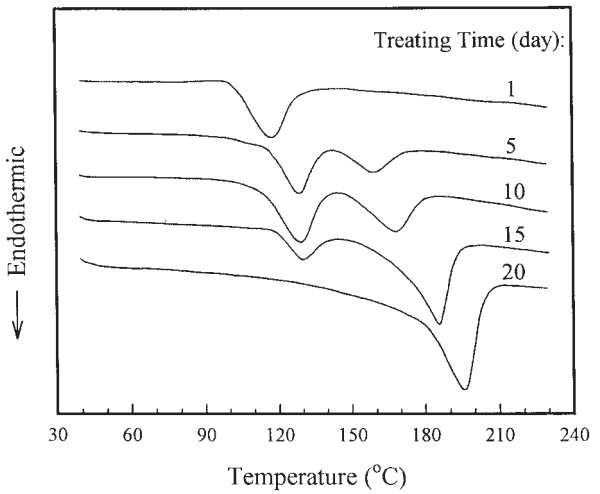

Figure 11 DSC thermograms of the IBF films, prepared from the powders iodinated with 1.0*N* I_2/KI aqueous solution, treated at 80°C for various periods of time.

The crystal types in the films isothermally treated at each temperature are summarized in Table II. In comparison with the samples treated at identical temperature, there are the less stable type of crystals in the film of the higher weight gain. This suggests that the film of the higher content of the I_2/KI obstructive to the crystallization should be treated at the higher temperature to form a more stable type of crystal. And the peaks in Figures 8–11 are generally weaker than those in Figures 2–5, which must be due to the higher content of I_2/KI as well.

In Figures 8 and 9, the peaks are too weak and broad to be concretely discussed. Merely, it is apparent that the temperature at which the peaks appear increases with the treating temperature and time. T_{max} and ΔH obtained from the endothermic peaks in Figures 10 and 11 are indicated in Table III.

In comparison with the samples treated for identical periods of time, T_{max} for the film treated at 80°C is higher than that for the film treated at 60°C, which is similar to the case of 0.2*N*. The dependence of ΔH on the treating time also indicates a similar tendency to the case of 0.2*N*: In the case of 60°C indicating a single peak, ΔH increases gradually with the treating time as it does in the case of 0.2*N*-80°C also indicating a single peak. In the case of 80°C, ΔH for the middle-temperature peak decreases while that for the higher-temperature peak increases, which is very similar to the case of 0.2*N*-60°C.

The X-ray diffractometer scans of the samples treated for 20 days are shown in Figure 12. The scans are more obscure and the diffraction peaks are still weaker than those in Figure 7, because these films contain more I_2/KI than the films from the powders iodinated with 0.2*N* I_2/KI solution. Therefore, a confident interpretation of the scans is difficult, but several signs on the scans are also helpful to support the preceding discussion on the thermograms. The films treated at 20°C and 40°C exhibit a very weak and broad peak at around $2\theta = 23°$, being related to the complex crystal. The film treated at 60°C exhibits a

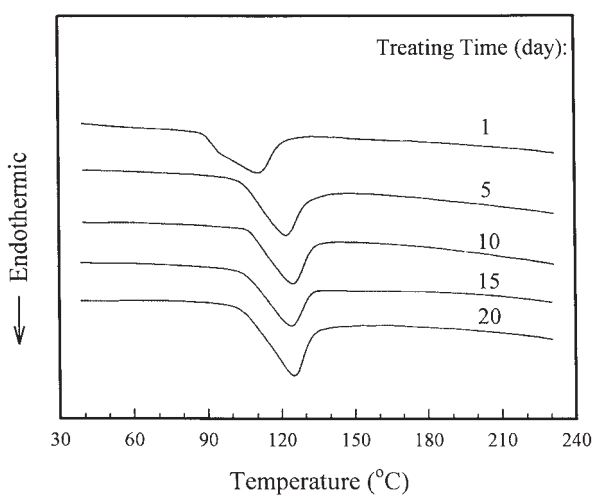

Figure 10 DSC thermograms of the IBF films, prepared from the powders iodinated with 1.0*N* I_2/KI aqueous solution, treated at 60°C for various periods of time.

TABLE II
Crystal Types in the Films Isothermally Treated at Each Temperature

Conc. of I_2/KI (weight gain)	Treating temp. (°C)			
	20	40	60	80
0.2N (194.9 %)	Complex + γ	Complex + γ	γ + α	α
1.0N (207.6 %)	Complex	Complex	γ	γ + α

TABLE III
Maximum Peak Temperature (T_{max}) and Energy of Fusion (ΔH) of the Endothermic Peaks for the Films Prepared from the Powders Iodinated with 1.0N I_2/KI Solution

Treating condition		Lower-temp. peak		Middle-temp. peak		Higher-temp. peak	
Temp. (°C)	Time (day)	T_{max} (°C)	ΔH ($\times 10^{-2}$J/g)	T_{max} (°C)	ΔH ($\times 10^{-2}$J/g)	T_{max} (°C)	ΔH ($\times 10^{-2}$J/g)
	1	—	—	110	34	—	—
	5	—	—	122	46	—	—
60	10	—	—	125	46	—	—
	15	—	—	126	49	—	—
	20	—	—	127	61	—	—
	1	—	—	118	46	—	—
	5	—	—	129	44	159	24
80	10	—	—	129	43	168	24
	15	—	—	131	24	185	61
	20	—	—	—	—	195	81

peak at around $2\theta = 22°$, being related to the γ-crystal ($2\theta = 21.8°$ and 22.7°). The film treated at 80°C exhibits peaks at around $2\theta = 21°$ and 24.5°, which are regarded to be slightly shifted from the peaks at $2\theta = 20.2°$ and 24.0°, being related to the α-crystal, and another peak at around 36°, also being related to the α-crystal.[15] These results can support the estimation by the thermograms that the IBF films treated for 20 days at 20 and 40°C, 60°C, and 80°C have the complex crystals, the γ-crystals, and the α-crystals, respectively. As mentioned above, the diffraction peak at around $2\theta = 25 \sim 26°$ is likely due to the KI crystals.

TG studies

Weight loss curves for the IBF films prepared from the powders iodinated with 0.2*N* and 1.0*N* I_2/KI aqueous solutions and treated for 20 days are shown in Figures 13 and 14, respectively.

It was presented in the previous paper[13] that the weight loss curve for the IBF film has two distinguishable weight loss zones, namely, the lower-temperature zone associated with volatilization of I_2 molecules isolated from I_5^- ions and the higher-temperature zone associated with decompositions of I_3^- ions and nylon 6 molecules.

In Figure 13 for the films prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution, the % weight loss in the lower-temperature zone is reduced with increased treating temperature. Such a reduction of the % weight loss may result from the volatilization of I_2 molecules isolated from I_5^- ions, which is the factor of the weight loss in the lower-temperature zone and which has previously oc-

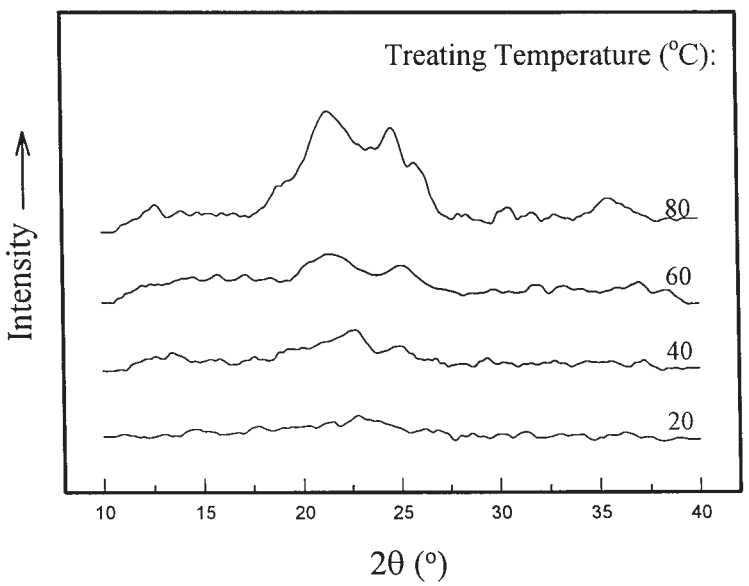

Figure 12 X-ray diffractometer scans of the IBF films, prepared from the powders iodinated with 1.0*N* I_2/KI aqueous solution, treated for 20 days at various temperatures.

curred through the long treating time of 20 days more abundantly with increased treating temperature. Besides, the I_2 molecules generated in the course of the TG performance may act as an oxidizer and reduce the degradation temperature of the nylon 6 molecules. It can actually be confirmed in several reports[12,13,15] as well as in Figure 12 that the degradation temperature of the iodinated nylon 6 was much lower than that of pure nylon 6. Therefore, with increased treating temperature, the reduction of the % weight loss in the lower-temperature zone, namely, the reduction of an amount of the I_2 molecules generated in the course of the TG performance, must cause the increase of the temperature of the higher-temperature zone closely connected with the degradation of nylon 6 molecules.

The variation tendency of the TG curves for the films from the powders iodinated with 1.0*N* I_2/KI solution, shown in Figure 14, is similar to that for the films from the powders iodinated with 0.2*N* I_2/KI solution. Merely, the variation amount is less, likely due to the higher content of I_5^- ions of the untreated film.

CONCLUSION

The DSC thermograms for the treated IBF films exhibit three groups of endothermic peaks at temperature ranges of 60 ~ 70°C, 105 ~ 120°C, and higher than 155°C, which may be associated with the melting of the complex crystal, the relaxed γ-crystal, and the relaxed α-crystals, respectively. The film containing less I_2/KI and treated at the higher temperature exhibits the peaks associated with the more stable type of crystal on its thermogram. In general, the maximum peak temperature increases with the treating temperature and time. With increased treating time on the occasion of being only a peak, ΔH increases gradually; on the occasion of being two peaks associated with the relaxed γ-crystal and α-crystal, the ΔH for the α-crystal increases while that for the γ-crystal decreases.

The TG curves generally have two temperature-zones of weight loss by the volatilization of the I_2 from I_5^- and the decompositions of I_3^- and nylon 6. With increased treating temperature, the % weight loss by the volatilization of I_2 decreases, and consequently the temperature of the weight loss by the decomposition of nylon 6 increases. The film containing more I_2/KI indicates the greater amount of the variations.

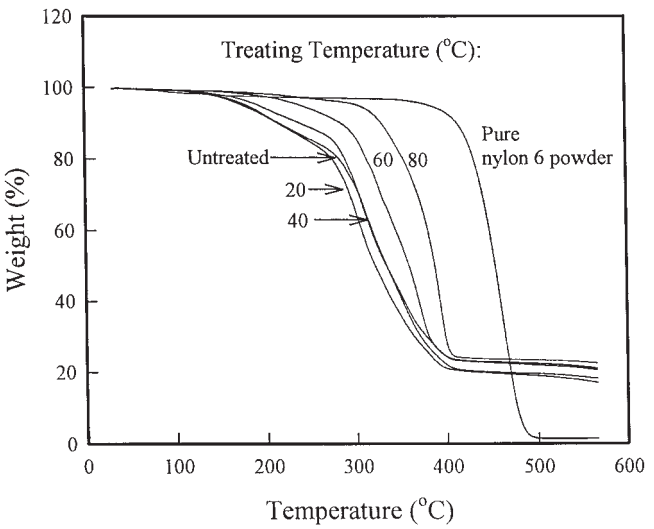

Figure 13 TG curves of the IBF films, prepared from the powders iodinated with 0.2*N* I_2/KI aqueous solution, treated at selected temperatures for 20 days.

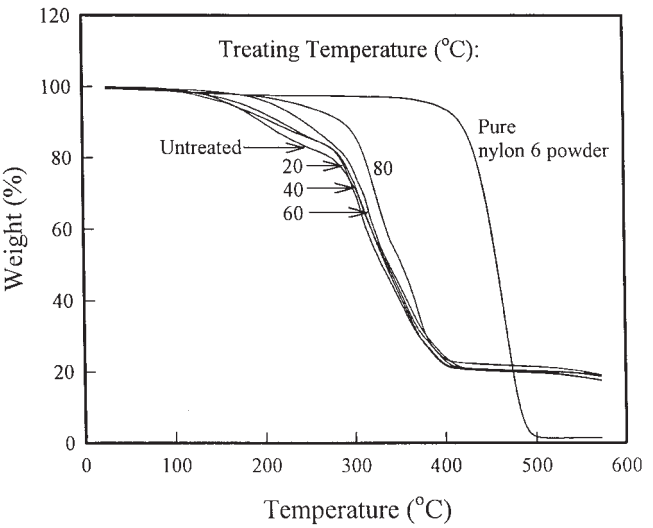

Figure 14 TG curves of the IBF films, prepared from the powders iodinated with 1.0*N* I_2/KI aqueous solution, treated at selected temperatures for 20 days.

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