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New Understanding on the Memory Effect of Crystallized *i*PP*

Xiang-yang Li[a], Zhe Ma[b], Feng-mei Su[a], Nan Tian[a], You-xin Ji[a], Jie Lu[a], Zhen Wang[a] and Liang-bin Li[a, c**]

[a] *National Synchrotron Radiation Laboratory, College of Nuclear Science and Technology, University of Science and Technology of China, Hefei 230026, China*

[b] *Department of Mechanical Engineering, Eindhoven University of Technology, P.O. Box 513, 5600MB, Eindhoven, The Netherlands*

[c] *Department o f Polymer Science and Engineering, CAS Key Lab of Soft Matter Chemistry, University of Science and Technology of China, Hefei 230026, China*

Abstract In this study, recovery processes of isotactic polypropylene (*i*PP) melted spherulites at 135 °C after melting at higher temperatures (170 °C–176 °C) were investigated with polarized optical microscopy and Fourier transform infrared spectroscopy. The recovery temperature was fixed to exclude the interference from heterogeneous nuclei. After melting at temperatures between 170 °C and 174 °C, the melted spherulite could recover back to the origin spherulite at low temperatures. Interestingly, a distinct infrared spectrum from *i*PP melt and crystal was observed in the early stage of recovery process after melting at low temperatures, where only IR bands resulting from short helices with 12 monomers or less can be seen, which indicates that the presence of crystal residues is not the necessary condition for the polymer memory effect. Avrami analysis further indicated that crystallization mainly took place in melted lamellae. After melting at higher temperatures, melted spherulite cannot recover. Based on above findings, it is proposed that the memory effect can be mainly ascribed to melted lamellae, during which crystalline order is lost but conformational order still exists. These conformational ordered segments formed aggregates, which can play as nucleation precursors at low temperatures.

Keywords: Memory effect; Isotactic polypropylene; Conformational ordering; Topological connectivity; Aggregate.

INTRODUCTION

Memory effect is a well-known phenomenon of polymers, which can be defined as the influence of previous crystalline morphology or chain orientation on subsequent crystallization kinetics when the previous thermal history is not erased completely. Based on the difference in origins, memory effects can be further classified into crystalline and orientational memory effects[1]. With the residual ordered structures, the nucleation density probably is increased by a few orders of magnitude[2–4], the growth rate of spherulites may be enhanced greatly[5–8], or polymer chains may orientate along the direction of previous flow field, exhibiting strong dependence on the previous thermal or processing histories.

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** Corresponding author: Liang-bin Li (李良彬), E-mail: lbli@ustc.edu.cn

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Classical memory effect experiments restrict the melting temperature into a narrow temperature range near or below the nominal melting point[6, 9, 10]. For example, Fillon *et al*. investigated the memory effect of *i*PP crystals with differential scanning calorimetry (DSC) and optical microscopy[10]. It was found that the memory effect existed only in a narrow range near or below the melting temperature (from 166 °C to 170 °C). Below 166 °C thick lamellae could be formed due to annealing, while above 170 °C non-isothermal crystallization kinetics was not affected by the change of fusion temperature. Note that strictly speaking the memory effect only existed between 166 °C and 168 °C, since only for samples melted in this range, a crystalline morphology similar to the previous one could be recovered at lower temperatures. Because the fusion temperatures employed in this study are slightly higher than the melting point, a viewpoint arises naturally that the surviving of a little of crystals after melting is the necessary condition for occurrence of memory effects. However, recent studies indicated that for polymers under shear flow[11–13], or some polymers like poly(ε-caprolactone) (PCL)[14], poly(ethylene oxide) (PEO)[15] or random copolymers[16], memory effects can be found after melting at temperatures much higher than the melting point, sometimes even above the equilibrium melting point. At such high temperatures, no crystal was found and the memory effect was often attributed to the existence of nucleating precursors[11, 13–16].

For *i*PP, the presence of crystalline residues seems to be a necessary condition for memory effect, but not for other polymers such as PEO, why? It may be ascribed to the difference in the crystallization kinetics. It is well known that at same supercoolings, PEO has faster crystallization kinetics than *i*PP, possibly due to its low molecular weight. When crystallized iPP is melted at higher temperatures, even some crystal residuals exist, the re-crystallization rate becomes slower. This allows the sample to crystallize at lower temperatures during a DSC cooling scanning, where a large number of heterogeneous nuclei were activated, which will undoubtedly obstruct the recovery process of melt spherulites.

The objective of this study is to study the crystallization process during recovery of melted spherulites. A spherulite was first melted at higher temperatures and then quenched to a lower temperature to recover. The recovery temperature is fixed to the previous crystallization temperature, in order to exclude the interference from heterogeneously nuclei at low temperatures as far as possible. Recovery processes of the melted spherulite were monitored with a polarized optical microscope (POM) and a Fourier transform infrared spectrometer (FTIR).

MATERIALS AND METHODS

The *i*PP sample used in this study has a weight-average molecular weight of 720 kg/mol, and a polydispersity of 4.8, which was kindly supplied by SABIC-Europe. An *i*PP film was sandwiched into two clean ZnSe windows for measurements of POM and FTIR. The use of same windows and hot stage for POM and FTIR measurements was to ensure same test temperatures.

For POM measurements, a large spherulite formed at 135 °C after removal of thermal history (220 °C for 5 min) was first subjected to melting at various temperatures from 166 °C to 176 °C for 5 min, and then it was quenched to 135 °C for recovery. The change of spherulite morphology during recovery was recorded every ten seconds using an Olympus polarized optical microscope with a charge coupled device (CCD). Note that the temperature has been carefully calibrated using a thermocouple.

For FTIR measurements, a completely crystallized *i*PP formed at 135 °C after removal of thermal history was first subjected to melting at various fusion temperatures from 166 °C to 176 °C for 5 min, and then quenched to 135 °C for isothermal crystallization. The change in IR spectrum during isothermal crystallization was recorded every 1 min with a Tensor 27 Fourier transform infrared spectrometer (FTIR). Note that before every measurement, isothermal crystallization processes at 135 °C after removal of thermal history were also recorded.

RESULTS

First of all, the melting process of *i*PP crystals was monitored with POM. Figure 1(a) shows morphological changes of a spherulite during increasing temperature. The spherulite was formed at 135 °C after erasing thermal history at 220 °C for 5 min. It was reported[6] that *i*PP crystals formed at temperatures lower than 145 °C only has one endotherm, and its peak temperature is around 165 °C. As seen in the Fig. 1(a), partial crystals start to melt at 166 °C. With further increasing the temperature to 168 °C, the spherulite disappears.

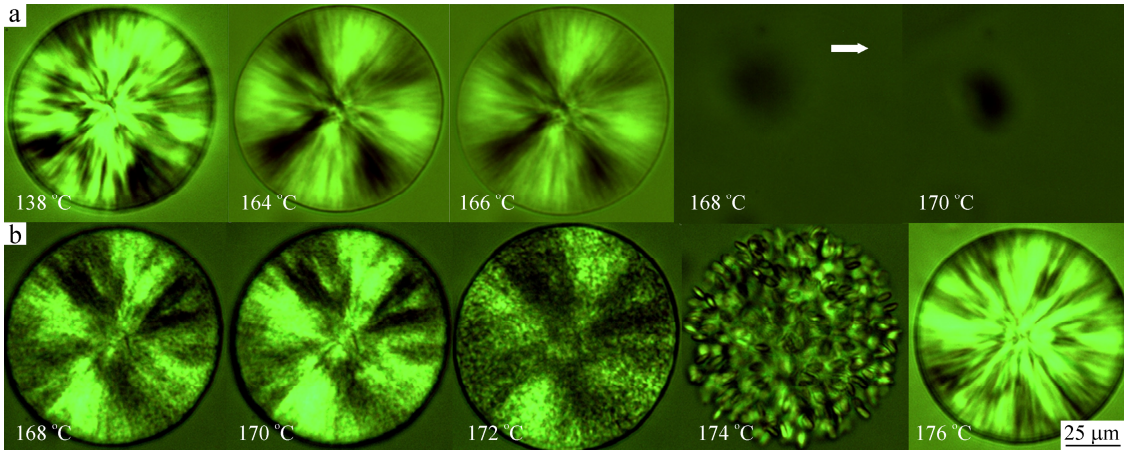


Fig. 1 (a) Melting process of a spherulite during increasing temperature from 135 °C to 170 °C, (b) spherulites recovered at 135 °C after being melted at different temperatures ranging from 168 °C to 176 °C for 5 min (The white arrow indicates the weak spherulitic boundary due to unfully melting.)

To identify the fusion temperature range where memory effect exists, the melted spherulite was cooled to 135 °C after holding at different temperatures for 5 min. After melting at 166 °C, the partially melted spherulite recovered quickly at 135 °C. This result can be understood straightforwardly. The un-melted crystals at 166 °C provide suitable substrate for polymer chains, reducing the nucleation barrier, thus secondary nucleation (or growth) could complete quickly at low temperatures. Surprisingly, when the spherulite disappears completely at 168 °C, the melted spherulite could also recover as shown in Fig. 1(b). Further increasing fusion temperature to 170 °C or 172 °C, the disappeared spherulite still can recover at the low temperature, though after melting at 172 °C the recovered spherulite did not have smooth surface as before. Figure 2(a) shows the recovery process of a spherulite after melting at 170 °C for 5 min. The outline of the spherulite first appears, and then the brightness of spherulite becomes stronger and stronger with time, indicating recrystallization.

When further increasing fusion temperature to 174 °C, though the melted spherulite could also recover at the low temperature, the nucleation density decreased significantly. The nuclei cannot fill in the overall area of the original spherulite. Thousands of nuclei were observed as seen in Fig. 2(b). More interestingly, these nuclei did not develop into spherulites but stripe-like crystals similar to single crystals[17]. When increasing fusion temperature to 176 °C, the spherulite cannot recover at 135 °C. Instead, a nucleus formed at the previous heterogeneously nucleating site and grew to a big spherulite as seen in Fig. 2(c). The growth rate of the spherulite was also measured. It was found that the growth rate was almost the same as that after removal of thermal history at 220 °C (not shown here), which further confirms that 176 °C was high enough to erase memory effect.

From above results, it can be concluded that *i*PP crystals could have memory effect in a broad temperature range from 166 °C to 174 °C. This melting temperature range found here is slightly higher than the previous results, which could be attributed to different procedures used[10, 14]. For previous studies, DSC was often employed, and the melted sample was cooled with a constant rate until complete crystallization. It is not hard to imagine that with increasing melting temperature, the recrystallizing rate becomes slower and slower. This

allows DSC to reach lower temperatures prior to crystallization, which may induce heterogeneous nucleating probably by a few orders of magnitude and cover the signal of memory effect. Differing from the DSC procedure, the melt was quenched to previous crystallization temperature. Only one active heterogeneous nucleating agent exists in the spherulite, which effectively reduces the interference from heterogeneous nucleating. Therefore, the real temperature range for the existence of memory effect can be identified.

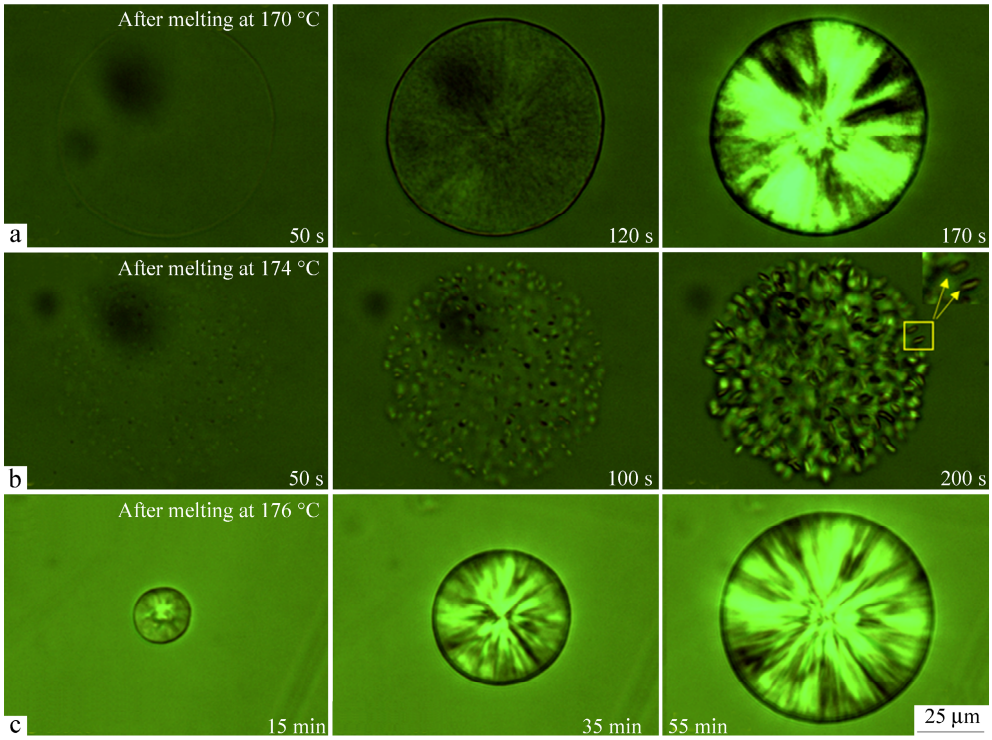


Fig. 2 Recovery processes of a melted spherulite at 135 °C after being melted at 170 °C (a), 174 °C (b) or 176 °C (c) for 5 min (Note that only one quarter of area in Fig. 1 was taken for observation easily.)

The recovery processes at 135 °C were also monitored by FTIR after melting of completely crystallized *i*PP at various fusion temperatures for 5 min. The crystallized *i*PP was obtained by isothermal crystallization at 135 °C for 80 min. Figure 3 shows changes in the IR spectrum and differential spectrum during isothermal crystallization at 135 °C after melting of *i*PP crystals at 170 °C for 5 min. For some regular bands, *i.e.* the bands at 808, 841, 900, 940, 998, 1040, 1102, 1165, 1220, 1303 and 1330 cm^{-1}, their peak areas increase with crystallization time (see Fig. 3b), which can be used to monitor the change in crystalline fraction during crystallization process. For some regular bands, *i.e.* the bands at 973 cm^{-1} and 1151 cm^{-1}, their peak areas decrease with crystallization time, which can be used to monitor the change in amorphous fraction. Note that the 973 cm^{-1} band actually consists of two bands located at 964 cm^{-1} and 976 cm^{-1} respectively, which can be clearly seen in Fig. 3(b). The two bands origin from two different helices, which contains 3 or 4 monomers, respectively[18]. In this study, the bands at 1151 cm^{-1} and 841 cm^{-1} are used to represent the amorphous and crystalline fractions, respectively. The crystallinity of the *i*PP sample (X_t) was determined using the following equation:

$$X_t = \frac{A_{841}}{A_{841} + A_{1151}} \tag{1}$$

where the normalized peak areas of bands at 841 cm^{-1} (A_{841}) and 1151 cm^{-1} (A_{1151}) are 0 and 1 at the beginning of the isothermal crystallization，while 1 and 0 after complete crystallization, respectively.

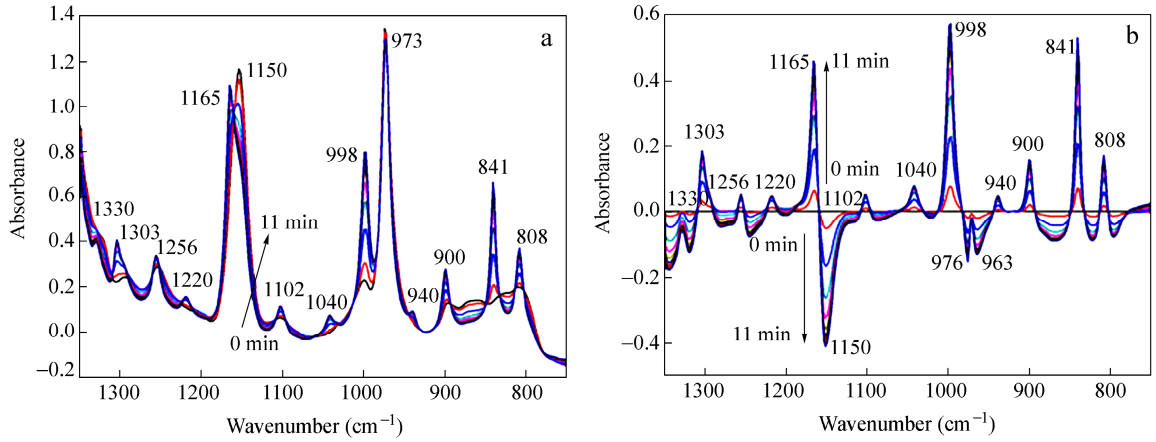


Fig. 3 Changes in the IR spectrum (a) and differential spectrum (b) during isothermal crystallization at 135 °C after melting of *i*PP crystals at 170 °C for 5 min

Figure 4(a) shows changes in crystallinity during recovery processes after melting of *i*PP crystals at 170, 172, 174 and 220 °C for 5 min. The recovery processes with fusion temperatures of 166 °C and 168 °C were also tried to be monitored. However, the recovery processes were too fast to be monitored. It can be seen that the time for complete recovery increases with increasing fusion temperature. When the fusion temperature reaches 176 °C, the time for complete recovery is almost the same as the time for completely crystallization after erasing thermal history at 220 °C. The isothermal crystallization processes were fitted with Avrami equation[19, 20]:

$$X_t = 1 - \exp(-kt^n) \tag{2}$$

or

$$\ln(-\ln(1 - X_t)) = \ln k + n \ln(t) \tag{3}$$

where X_t , k, and n are the normalized crystalline mass fraction, crystallization rate constant and Avrami exponent, respectively. The Avrami exponent represents the dimensionality of the growing crystals (a value of 3 corresponds to unrestricted three-dimensional crystal growth, a value of 2 to two-dimensional crystal growth) as well as the time dependence of nucleation (0 corresponds to instantaneous, athermal nucleation and 1 to purely sporadic, thermal nucleation). The fitting processes and the fitting results are shown in Figs. 4(b) and 4(c), respectively. Note here that all data have been used for the fitting except the first and the last ones, since both the data with X_t = 1 and with t = 0 lead to negative infinity in one side of Eq. (3). It can be seen that the crystallization rate becomes lower with increasing fusion temperature, and the Avrami exponent increases with increasing fusion temperature. When fusion temperature reaches 176 °C, the crystallization rate and Avrami exponent have the same values as those after removal of thermal history at 220 °C. The Avrami exponent of 3 indicates that nucleation occurred in bulk not on the surface. When fusion temperature is below 176 °C, the Avrami exponents were around 2. Because the nucleation process during recovery process is commonly assumed to be athermal, the Avrami exponent of 2 only can be attributed to two-dimensional growth. With two-dimensional growth and observed stripe-like crystals, we suggest that the recovery process presumably proceeds within isolated melted lamellae, which only have two growth dimensions. The classical viewpoint that the recovery of spherulite is achieved through growth of thousands of smaller spherulites is questionable, since the Avrami exponents obtained by non-isothermal crystallization process from other researchers[6] or our isothermal crystallization are around 2, which is impossible for three-dimensional growth. Li *et al.*[21] observed

morphological change on a lamellar level using scanning electron microscope. It was found that recovery of radial parent lamellae was responsible for the recovered spherulite, which supports our expectations.

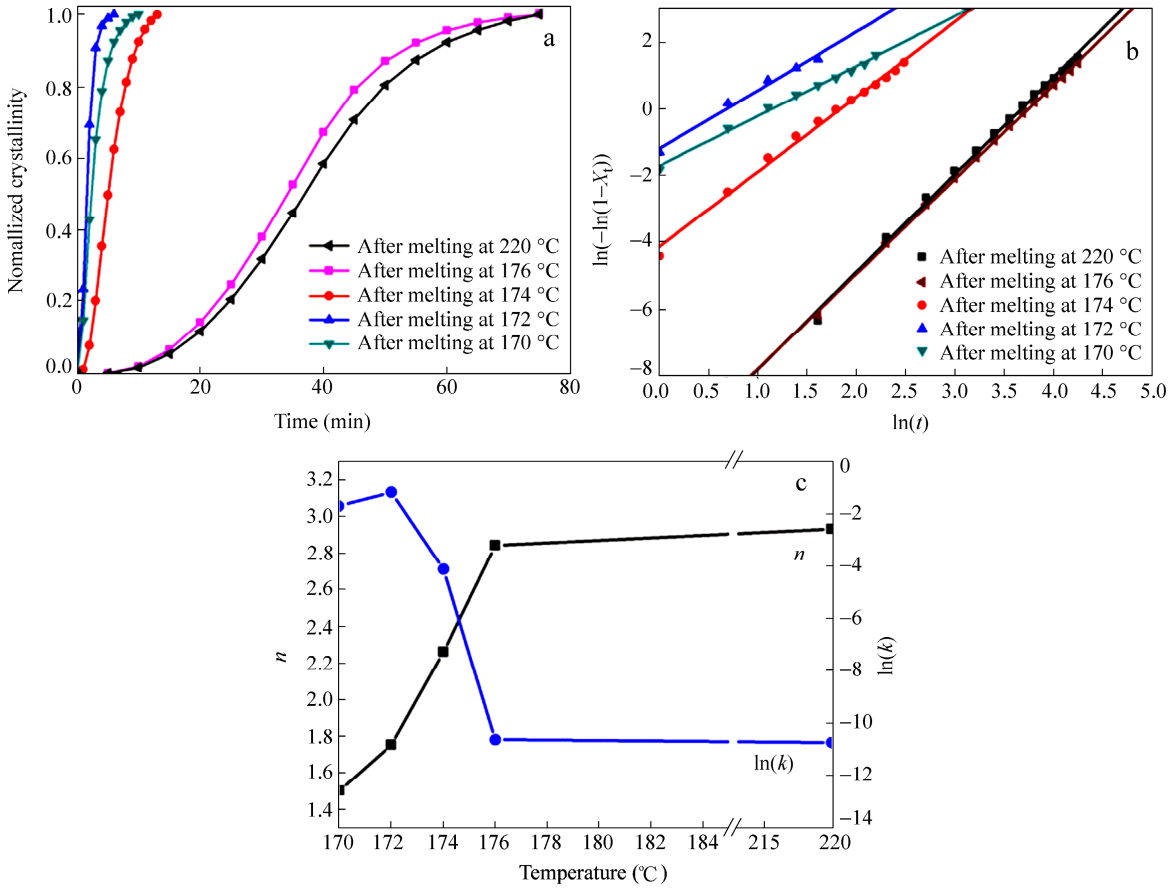


Fig. 4 (a) Changes in crystallinity during isothermal crystallization at 135 °C after melting of *i*PP crystals at 170, 172, 174 and 220 °C for 5 min; (b) Fitting processes with Avrami equation and (c) Temperature dependence of Avrami exponent and crystallization constant

In order to further clarify the origin of memory effect, we employed a special differential spectrum to check in the beginning of isothermal crystallization whether some ordered structures existed in the melts with different thermal histories. The differential spectrum was obtained through the first spectrum of isothermal crystallization at 135 °C after melting at a lower temperature (170, 174 and 176 °C) minus that after removal of thermal history at 220 °C (see Figs. 5a–5c). Note that the latter isothermal crystallization process at 135 °C is in fact the sample preparation process for the former one (see experimental section). Because the same film and crystallization temperature are used, the influence of thickness and temperature on the differential spectrum can be eliminated through subtracting. The differential spectrum could give weak signals of ordered structures possibly existing in the melts with low-temperature melting.

From Figs. 5(a)–5(c), it can be seen that two original spectra in every figure almost overlap each other. This indicates that this procedure indeed can eliminate the spectral differences caused by film thickness or temperature. In addition, no bands at 841, 940, 1041, 1220 and 1303 cm^{-1} can be seen from the original spectra, indicating that polymer was still in the induction time. After subtracting, some weak differences appear, and the differences depend on the fusion temperature. For the melts with heating at 174 °C and 176 °C, the differences are located at 973, 1151, 1320 and 1340 cm^{-1}, which probably is due to weak difference of amorphous fractions. For the melt with heating at 170 °C, except from the difference in amorphous bands, the differences also can be

found at 808, 841, 900, 998, 1033 and 1102 cm^{-1}. Part of the above bands, such as 808, 900, 998 and 1102 cm^{-1}, origins from helices with 6–10 monomers. They are regarded as conformational bands, which still exist after melting of crystals. This indicates that the melt with heating at 170 °C contains rich conformational ordered segments. More interestingly, the differential spectrum contains the 841 cm^{-1} band, which origins from helices with 12 monomers. According to previous studies[17, 22, 23], the appearance of the band at 841 cm^{-1} could induce the occurrence of isotropic-liquid crystal transition, despite no further evidence of existence of liquid crystal at present. Need to note that only when bands at 940, 1220, 1303 and 1330 cm^{-1} appear, which origins from helices with more monomers than 12, crystals could form. In the differential spectrum, no such bands can be found. Overall, IR spectrum is distinct from those of *i*PP crystal and melt, which indicates that a mesomorphic state/phase between crystal and amorphous may form. Similar differential spectrum was also found in the melt with melting at 172 °C. The 841 cm^{-1} band still can be observed but its intensity becomes very weak. In the melt with heating at 174 °C, the 841 cm^{-1} band is absent, even for the bands at 800, 900, 998 and 1102 cm^{-1}. This indicates that the presence of crystal residues is not necessary condition for the memory effect of polymer.

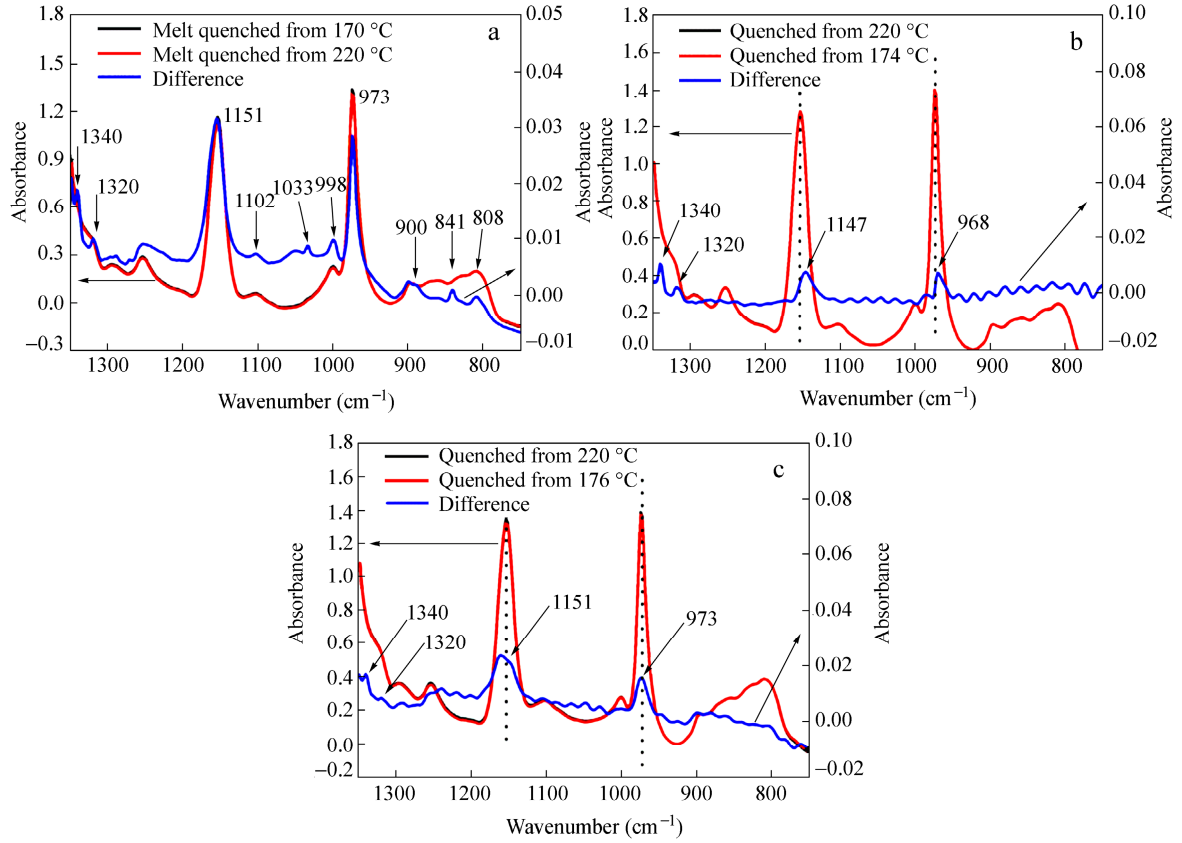


Fig. 5 First IR spectra of *i*PP melt during isothermal crystallization at 135 °C after melting at 170 °C (a), 174 °C (b) and 176 °C (c) for 5 min
First spectra of *i*PP melts during isothermal crystallization at 135 °C after melting at 220 °C for 5 min and differential spectra with that after melting at lower temperatures are also given in the plots.

For further comparison, the above three differential spectra are given in Fig. 6. IR spectrum from *i*PP crystal obtained by melt crystallization after removal of thermal history is also shown. It can be seen the melt quenched from 170 °C has different spectral nature from that quenched from 220 °C. Only for the melt quenched from 220 °C the bands at 1220, 1303 and 1330 cm^{-1} exist, though both melts have almost same peak area at 841 cm^{-1}. In the melt quenched from 170 °C, rich conformational bands with short helices such as the bands at

808, 841 and 900 cm^{-1} are observed. For the melt with melting at 174 °C, no conformational ordering bands can be observed in the differential spectrum. Only peak shifts can be found in the bands at 1151 cm^{-1} and 973 cm^{-1}. The origin of peak shifts is not clear, which needs to be further investigated in the future.

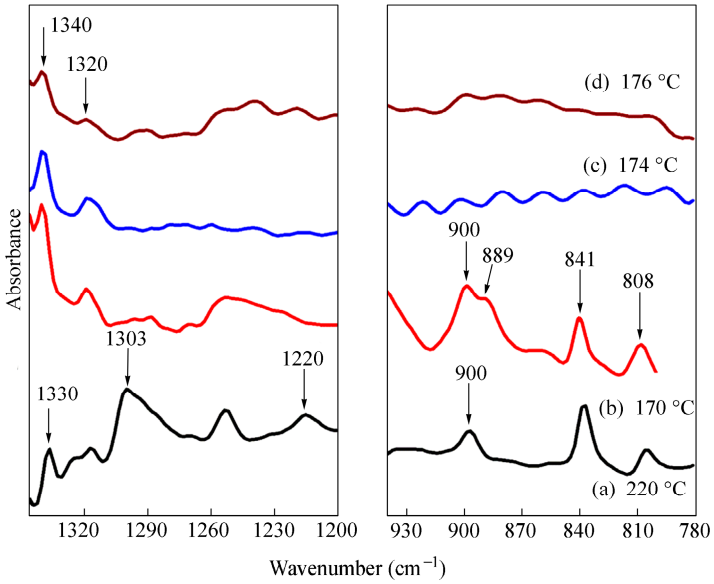


Fig. 6 Differential spectra during induction time after melting of completely crystallized *i*PP at 220 °C (a), 170 °C (b), 174 °C (c) and 176 °C (d)

DISCUSSION

From the above results, it can be clearly seen that the recovery process of melted spherulites does not definitely require the presence of crystal residues. At higher melting temperatures, *i*PP molecules still had memory on the previous crystalline information, where crystalline order was lost but conformational order existed. Avrami analysis indicates that the crystallization mainly took place with melted lamellae. If no crystalline residues are present in the melted spherulite, how does the melted spherulite recover?

Before discussion on the recovery mechanism, let us image the influence of melting temperature on the spherulitic morphology on the lamella. From Fig. 1, it can be seen that after melting at 170 °C and 172 °C whole melted spherulite can recover, implying that no polymer chains run out of the spherulite. It is well known that the density of crystal is higher than that of amorphous fractions, thus the density of spherulite is also higher than that of its surrounding melt, though the spherulite commonly contains some amorphous fractions. How to let more polymer chains fill into a melted spherulite? The most possible method is to let some polymer chain segments arranged paralleling to each other. Clearly, if no attractive interactions exist among these segments, they will disappear due to entanglement. Therefore, there should exist attractive interactions. Under these interactions, these segments form ordered structures. In the melt with heating at 170 and 172 °C, bands at 1220, 1303, and 1330 cm^{-1} were absent, indicating the absence of crystals. Therefore, these ordered structures are not crystalline. The only possibility is that these ordered structures formed through aggregation of short helices. The presence of the band at 841 cm^{-1} can be regarded as the hint of ordered structures.

With further increasing the fusion temperature to 174 °C, a great number of aggregates may melt. The aggregates consisting of chain segments arranged parallel to each other disappeared quickly due to entanglement. The reptation time at 180 °C was estimated through small-amplitude oscillation shear measurements using Physica MCR 301 rotational rheometer, which was found to be 0.6 s. Melting for 5 min at 174 °C is enough for the entanglement. The configurational entropy increases with re-entanglment, which increases volume exclusion interactions. When the interactions are big enough against the resistance from the network, some polymer chains

run out from the network. This decreases the network density and favors the relaxation of ordered structures.

As is well known, an important difference between polymer and small molecules in crystallization is topological connectivity of polymer, that is, a polymer chain could traverse different lamellae and thus connect them together forming a small network. The higher the molecular weight, the big the network. Therefore, the higher molecular weight of polymer chains has stronger memory, as reported by some researchers[11]. Within the network, the chain mobility was reduced significantly, preventing the relaxation of aggregation into random coil.

Based on the above results, possible recovery routes of melted spherulites after melting at 170 °C (a) and 174 °C (b) are described in Fig. 7. During melting at 170 °C, lamellae melt, however, the polymer chain does not relax into random coils. Some short helices may exist, which forms aggregates. At low temperatures, they can play as nucleation sites. The density of nuclei depends on the melt temperature at same melting time, which decreases significantly with increasing fusion temperature[1]. In the melt with heating at low temperatures, the nucleation is dominant during crystallization, while the crystal growth becomes dominant after melting at higher fusion temperatures. At low temperatures, the homogeneous nucleation contributes 1 to the Avrami exponent, while the growth of crystal contributes 0, leading to the overall Avrami exponent of 1. With increasing the fusion temperature, the contribution of growth to the overall crystallization kinetics becomes big, resulting in the increase of Avrami exponent. During increasing fusion temperature to 174 °C, a great amount of subsidiary lamellae melted and relaxed to random coil. This increases the configurational entropy and thus the volume exclusion interaction, leading to running out of the melted spherulite. In the melt, only melted dominant lamellae along the diameter exist (see Fig. 7b), which play as nucleation precursor at low temperatures and induce the formation of single crystals as reported recently[24]. At this moment, the contribution of crystal growth to the overall crystallization kinetics becomes dominant. The single crystal has two growth directions, which leads to Avrami exponents around 2.

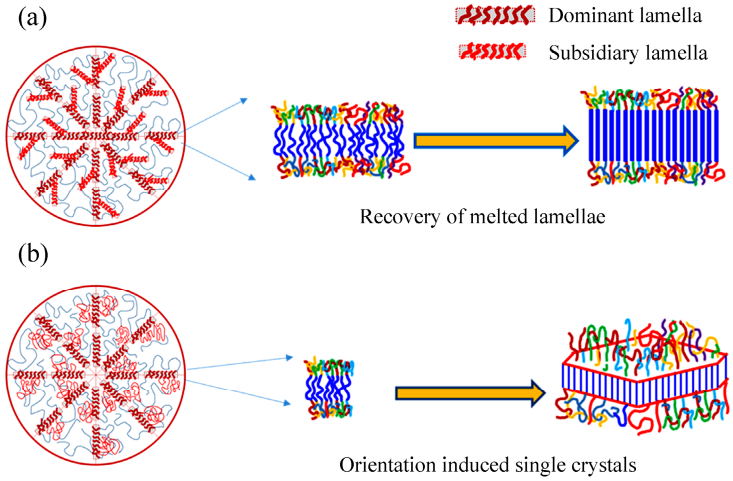


Fig. 7 Possible recovery routes of melted spherulite at 170 °C (a) and 174 °C (b)

CONCLUSIONS

In this study, recovery processes of melted spherulites after melting at high melting temperatures were investigated with POM and FTIR. It was found that the presence of crystal residues was not the necessary condition for polymer memory effect. In the memorized melts with heating at 170 °C and 172 °C, only conformational order existed and crystal was absent. Avrami analysis indicated that the re-crystallization mainly occurs within the melted lamellae. Based on the above findings, it was proposed that the lamellae melted but did not relax into random coils, which existed in the form of aggregates and played as nucleation precursors at low temperatures.

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