

# The Effect of $\gamma$ -Irradiation on Thermal Strain of High Strength Polyethylene Fiber at Low Temperature

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**ABSTRACT:** To understand the contribution to negative thermal expansion by the length of the molecular chains in high-strength ultra-high-molecular-weight polyethylene (UHMW-PE) fiber, the thermal expansion coefficient in the range of low temperature was investigated for high-strength UHMW-PE fiber (Toyobo, Dyneema®; hereinafter abbreviated to DF), irradiated by  $\gamma$ -rays ( $\gamma$ -rays treatment) that induce the molecular scission. The molecular weight of DF decreased by  $\gamma$ -ray treatment. X-ray diffraction behavior did not change by  $\gamma$ -ray treatment. The melting behavior observed by DSC showed the main chain scission of DF by  $\gamma$ -ray treatment. The DFs, with and without  $\gamma$ -ray treatment, expand by cooling down (negative thermal expansion). The

change of negative thermal expansion of DF by  $\gamma$ -ray treatment was small. It is suggested that negative thermal expansion does not change by only the molecular chain scission. These results suggested that the effect of negative thermal expansion of DF in the temperature range from 213 to 303 K by the molecular chain scissions is small and that the length of extended molecular chains contributes to a negative thermal expansion a little. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 204–209, 2006

**Key words:** negative thermal expansion; thermal strain; ultra-high-molecular-weight high-strength polyethylene fiber;  $\gamma$ -rays; scission

## INTRODUCTION

Recently, the importance of polymeric materials as electrical insulating or structural materials has been recognized in those areas of cryogenic engineering that require superconducting technology, such as superconducting power equipment, magnetic levitated transportation, medical diagnosis equipment, nuclear fusion reactors, and so on. To apply polymeric materials to these areas, their thermal properties, and in particular their coefficient of linear expansion and their thermal conductivity, have been important parameters to study. For example, high-strength polyethylene (PE) fiber (Toyobo, Dyneema®; hereinafter abbreviated to DF) possesses a negative coefficient of linear expansion toward the direction of the fiber axis.<sup>1–3</sup> Typical applications for these materials are as coil bobbins<sup>4–14</sup> and spacers<sup>8,13,15,16</sup> for superconductors and package of Fiber Bragg Grating for optical filter.<sup>17,18</sup> Those materials are desired not only as thermal insulators but also as electrical insulating thermal conductors for thermal conductivity.<sup>16,19</sup>

It is known from X-ray studies that the thermal expansion coefficient of polymer crystals, for example, PE in the direction of the chain axis measured, is negative for most if not all polymers<sup>20–22</sup> The negative thermal expansion is explained by “Linear Chain Model” and so on.<sup>23–25</sup> Therefore, highly oriented and highly crystallized PE shows a negative thermal expansion in the oriented direction. This thermal expansion can be treated by a mechanical model composed of a crystal part showing a negative thermal expansion and an amorphous part showing a positive thermal expansion (Takayanagi model).<sup>22,26</sup>

The DF is a high-crystallinity and high-strength fiber made from ultra-high-molecular-weight PE (UHMW-PE).<sup>26–29</sup> As aforementioned, previous reports showed that DF has a negative thermal expansion coefficient.<sup>1–3</sup> A previous report elucidated that the negative thermal expansion of DF is explained by the serial-parallel mechanical model composed of crystal and amorphous region.<sup>3</sup> In the case of DF, it is considered that the negative thermal expansion of extended molecular chains in continuous crystal part in chain axis contributes to that of DF in the fiber direction.<sup>3</sup> Meanwhile, the effect on negative thermal expansion of the length of extended molecular chains by using the UHMW-PE has not been investigated systematically. Therefore, the relation between the

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negative thermal expansion and the molecular length of DF is investigated in this work.

It is well known that PE undergoes main chain scission by irradiation with  $\gamma$ -rays in the presence of oxygen.<sup>31,32</sup> In this article, we report the thermal expansion coefficients of DFs affected by main chain scission and induced by irradiation with  $\gamma$ -rays, for investigation of the relation between the thermal expansion coefficient and the length of main chain, to clarify the effect of negative thermal expansion on the chain length of DF.

## EXPERIMENTAL

### Sample

In this work, thermal expansion coefficients of DF and those irradiated with  $\gamma$ -rays ( $\gamma$ -ray treatment) were measured in the range of low temperatures. The used DF was Dyneema®, SK-60 with 1320dtex (Toyobo). Irradiation was carried out with Co-60  $\gamma$ -rays at ISIR (Sanken), Osaka University, at an ambient temperature and in air. The absorbed dose rates were 33.3 and 83.3 kGy/h. Irradiation time was 6 h. The total absorbed doses were 0.2 and 0.5 MGy, respectively. These are hereinafter abbreviated to DF (0.2 MGy) and DF (0.5 MGy). The DF without  $\gamma$ -rays treatment is abbreviated to DF (Blank).

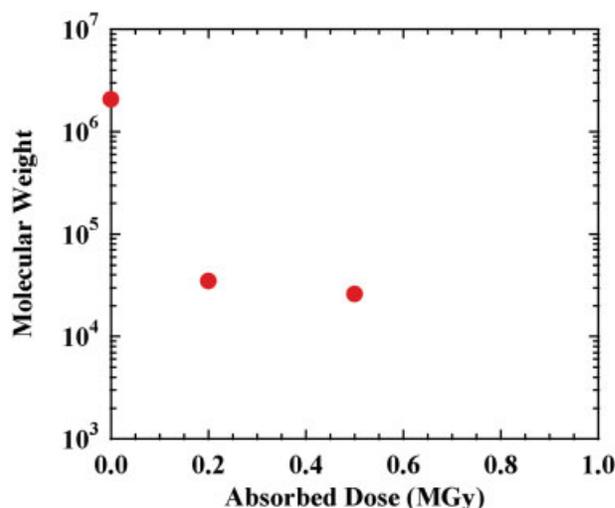
### Measurements

#### Molecular weight

To observe the relative decreasing of the length of molecular chains by irradiation with  $\gamma$ -rays, the viscosity-average molecular weight of DFs, with and without  $\gamma$ -ray treatment, were measured by the viscosity method, using decalin solution. The DFs, with and without  $\gamma$ -ray treatment, were dissolved in decalin with a concentration of 0.0045 g/dL. The relative viscosities at 408 K were measured by using Ubbelohde viscometers in a thermostat. This method was reported in elsewhere.<sup>33</sup>

#### Crystallinity and orientation

X-ray diffraction and melting behavior by differential scanning calorimetry (DSC) were observed, to investigate the relative change of crystallinity of DFs by  $\gamma$ -ray treatment. A Rigaku RU-200 (40 kV  $\times$  100 mA) was used with X-ray diffraction on Ni-filtered Cu K $\alpha$  ( $\lambda = 0.1548$  nm). The distributions of (110) and (200) diffraction spots in the equatorial direction were observed. DSC measurements of DFs were carried out on a Perkin-Elmer DSC-7. An empty sample pan was taken as the reference sample. The samples were weighted at 2 mg, and the melting experiments were



**Figure 1** Dependence of molecular weight of DF on absorbed dose irradiated by  $\gamma$ -rays. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

made in the temperature range from 323 to 523 K at a heating rate of 1°/min.

#### Thermal expansion coefficients

Thermal expansion coefficients of DFs were measured by thermomechanical analysis (TMA) on tensile mode. TMA measurements were carried on a MAC Science TMA 400S. Samples were run in TMA in the temperature range from 213 to 303 K, at a scanning rate of 5°/min. Sample length was 20 mm. The loading was 240 g/mm<sup>2</sup>.

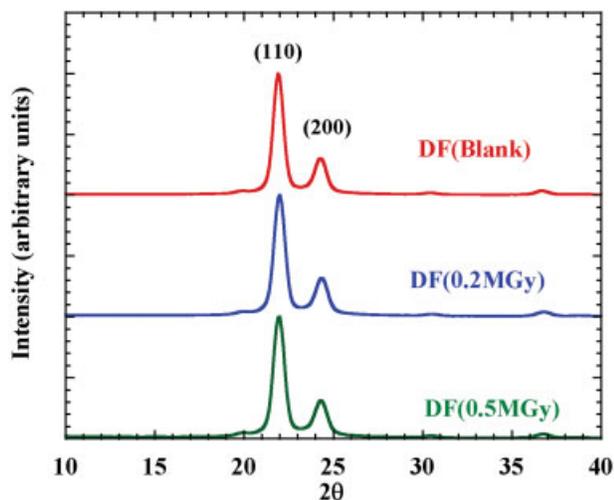
## RESULTS AND DISCUSSION

### Molecular weight

The relation between the viscosity-average molecular weight of DF and absorbed dose is shown in Figure 1. The measured molecular weight of DF (Blank) was  $2.0 \times 10^6$ . It decreases to  $3.5 \times 10^4$  by 0.2 MGy irradiation and to  $2.6 \times 10^4$  by 0.5 MGy irradiation, respectively. This result shows what the main chain scission of DF undergoes by the  $\gamma$ -ray treatment.

In the irradiation effects of PE in air, the free radicals of alkyl type are initially produced by irradiation, and the radicals combine with oxygen molecules into peroxy radicals. The peroxy radicals eliminate the hydrogen from the neighboring PE molecular chains, and hydroperoxides are produced. The decompositions of the hydroperoxides induce main chain scission of PE.

Therefore, the irradiation effects of PE in air depend on the diffusion of oxygen.<sup>31,32</sup> It is considered that oxygen invades into the amorphous region more easily than into the crystal region in DF. Therefore, it is



**Figure 2** X-ray diffraction profiles of DFs irradiated and unirradiated with  $\gamma$ -rays. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

considered that the probability of main chain scission is higher in amorphous region than in crystal region in DF.

#### X-ray diffraction measurements

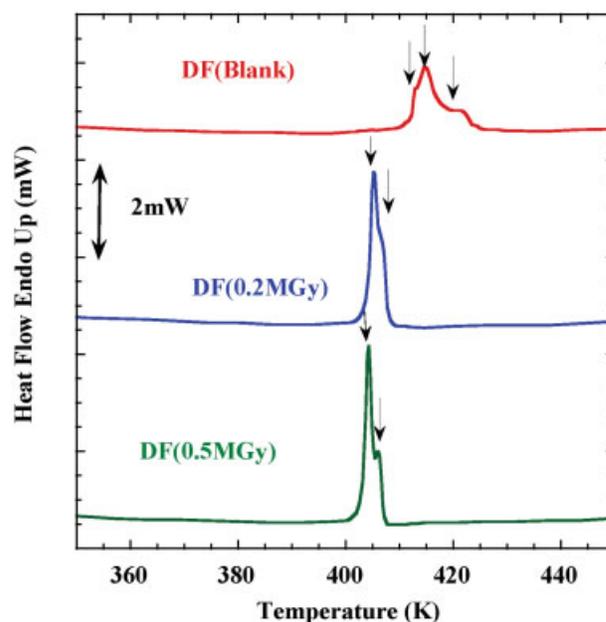
X-ray diffraction profiles of DFs in the equatorial direction are shown in Figure 2. The assignments of the reflections are shown in the figure. The changes of those reflection peaks by  $\gamma$ -ray treatment are investigated. The width, shapes, intensities, and reflection angles of those peaks do not change by  $\gamma$ -ray treatment. This result suggests that the crystal size in (110) and (200) directions do not change. From the independence of X-ray behavior with  $\gamma$ -ray treatment, it is considered that the crystal of DF is hardly destructed by  $\gamma$ -rays. The crosslinking reactions dominate in the case of irradiation of PE in the vacuum. It is reported that X-ray diffraction profile in equatorial direction does not change the irradiation of  $\gamma$ -rays, in which the absorbed dose is less than 0.25 MGy in vacuum<sup>34</sup> as like the case of this work.

#### Melting behavior

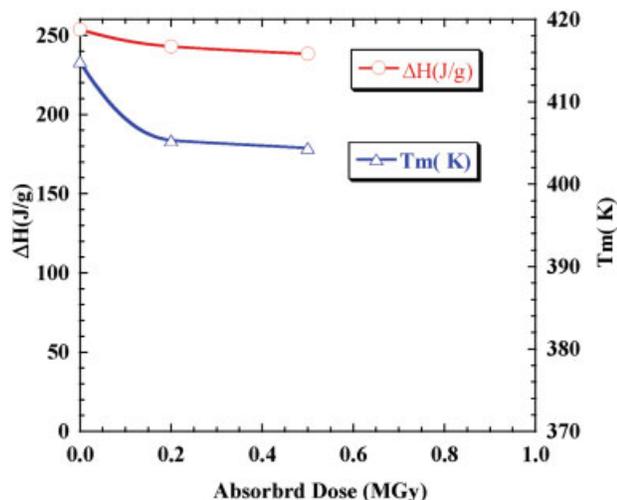
The melting behaviors of DFs observed by DSC are shown in Figure 3. DF (Blank) shows three broad melting peaks in the temperature range from 403 to 423 K. The melting behaviors of high-strength PE fiber as like DF are reported in other papers.<sup>35–37</sup> Although the assignments of the peaks in melting process of high-strength PE fiber are not clarified, it is reported that UHMW-PE molecular chains extended by the tensile stress show the melting behaviors<sup>35–37</sup> as like that of DF (Blank).

In the next, the radiation effect of melting behavior is investigated. The melting peaks of DF (0.2 MGy) and DF (0.5 MGy) are more sharp than that of DF (Blank). And the number of peaks decreases from three to two by  $\gamma$ -ray treatment. This change suggests the decrease of restraint on molecular chains in crystal region with  $\gamma$ -ray treatment. It is considered that the molecular chains in amorphous region are extended, and are restrained by orientation in the drawing. And it is also considered that there are interactions between the extended molecular chains in crystal region and those in the surrounding amorphous regions or the neighboring crystal regions. From the X-ray diffraction in the previous section, the crystal size is inferred to decrease not so much by  $\gamma$ -ray treatment. But it is considered that the molecular chains in amorphous region become more movable by main chain scission with  $\gamma$ -ray treatment and that the interactions of molecular chains between the crystal and amorphous regions decrease very much. The sharpening of the peaks in melting behavior is inferred to be caused by decreasing those interactions by main chain scissions by  $\gamma$ -rays.

The melting enthalpies ( $\Delta H$ ) and melting temperatures ( $T_m$ ) of DF, with and without  $\gamma$ -ray treatment, are shown in Figure 4. Both of them decrease by  $\gamma$ -ray treatment even though X-ray diffraction behavior does not change. The differences of  $\Delta H$  and  $T_m$  of DF, between with and without  $\gamma$ -ray treatment, are remarkable. On the other hand, the differences of  $\Delta H$  and  $T_m$  are small, with an absorbed dose between 0.2 and 0.5 MGy. It is considered that the decrease of  $\Delta H$  and  $T_m$  with  $\gamma$ -ray treatment is caused by the decrease



**Figure 3** DSC melting curves of DFs. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 4** Melting enthalpy ( $\Delta H$ ) and temperature ( $T_m$ ) of DFs. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

of interactions described in the previous paragraph by main chain scission. The detailed discussion about structural analysis will be necessary to clarify the mechanism of the melting behavior mentioned earlier.

The melting behavior observed by DSC also suggested the main chain scission in DF by  $\gamma$ -ray treatment.

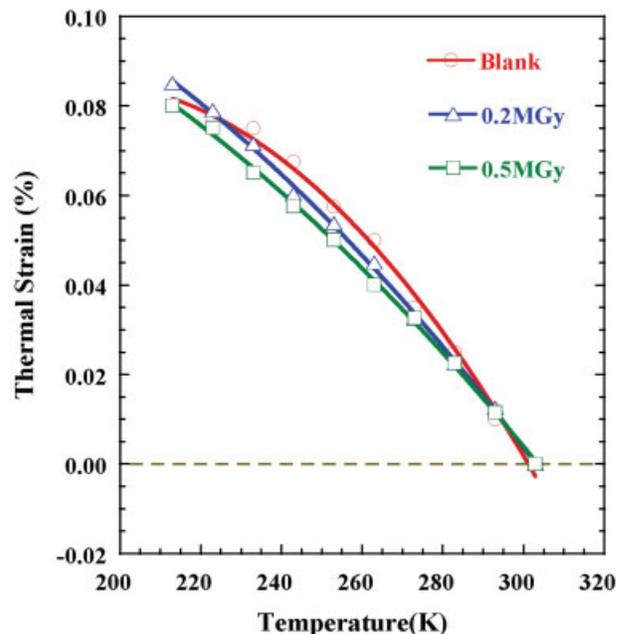
From the results of molecular weights, X-ray diffraction, and melting behavior, main chain scissions occurred by  $\gamma$ -ray treatment, with an absorbed dose of 0.2 and 0.5 MGy, though the crystal region did not change distinctly.

### Thermal expansion coefficients

Thermal strains of DFs in fiber direction measured by TMA are shown in Figure 5. All of them expand by cooling down (negative thermal expansion).

We investigate the effect of irradiation with  $\gamma$ -rays to thermal strain in the following. Thermal strain does not change by  $\gamma$ -ray treatment mostly. Although the temperature dependence of thermal strain becomes slightly straight by  $\gamma$ -ray treatment, the change is very small. Comparing DF (0.5 MGy) with DF (0.2 MGy), the inclination of DF (0.5 MGy) is a little smaller than that of DF (0.2 MGy) and those show similar behaviors. The average thermal expansion coefficients of DFs are estimated by the inclinations of the thermal strains of DFs, shown in Figure 5, in the range from 250 to 300 K. All samples show an almost straight line in this temperature range.

The relation between the negative thermal expansion and absorbed dose of  $\gamma$ -rays is discussed here. As aforementioned, those  $\gamma$ -ray treatments induced only a decrease of molecular weight, that is to say, the reduction of length of molecular chain. Crystal structure did not change by  $\gamma$ -ray treatment in the X-ray



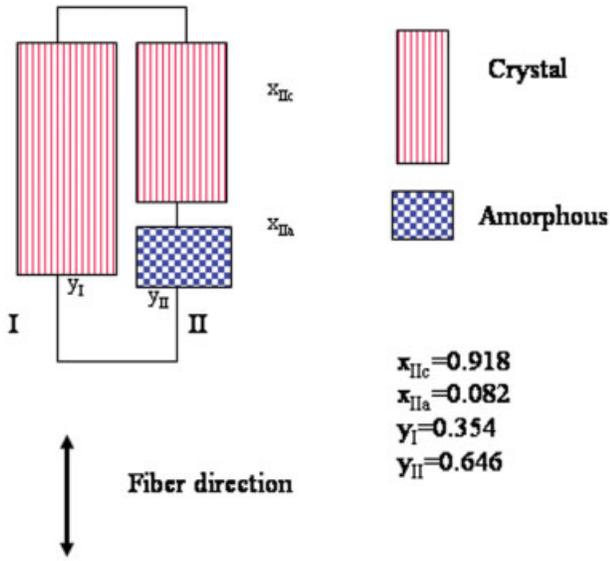
**Figure 5** Thermal strain of DFs with and without  $\gamma$ -rays treatment in fiber direction. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

diffraction measurement. The average thermal expansion coefficients, absorbed dose with  $\gamma$ -rays, and average molecular weight of DFs, with and without  $\gamma$ -rays treatment, are shown in Table I. Although negative expansion shows the tendency to decrease a little by irradiation, that is, the molecular chain scission, the change is small.

In the next, the relation between thermal expansion coefficient and molecular chain scission is discussed. Some kinds of physical properties of DF in the fiber direction are explained by the serial-parallel mechanical model composed of a crystal region and amorphous region (Takayanagi model). In this model, DF is composed of the following two parts by parallel combination. One of them is the continuous crystal region (hereinafter Part I) and the other is the series combination part of crystal and amorphous region (hereinafter Part II). In previous papers,<sup>3</sup> it is reported that thermal expansion coefficient of DF is explained by

**TABLE I**  
Molecular Weights and Thermal Expansion Coefficients of DFs

Sample	Absorbed dose (MGy)	Average molecular weight ( $10^4$ )	Thermal expansion coefficient at 253–303 K ( $10^{-6}/K$ )
DF (Blank)	0	200	–12.0
DF (0.2MGy)	0.2	3.5	–11.4
DF (0.5MGy)	0.5	2.6	–10.5



**Figure 6** Schematic diagram showing the structure of DF by a mechanical model. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

this serial-parallel mechanical model (Takayanagi Model). The schematic diagram, showing the structure of DF by the mechanical model used in the previous papers,<sup>3</sup> is shown in Figure 6. The

thermal expansion coefficients of DFs are estimated by the serial-parallel mechanical model using the following formulas:

$$\alpha_I = \alpha_c \quad (1)$$

$$\alpha_{II} = X_{IIc}\alpha_c + X_{IIa}\alpha_a \quad (2)$$

$$\alpha = [y_I E_I \alpha_I + y_{II} E_{II} \alpha_{II}] / [y_I E_I + y_{II} E_{II}] \quad (3)$$

$$E = [y_I E_I + y_{II} E_{II}] \quad (4)$$

$$E_I = E_c \quad (5)$$

$$E_{II} = E_c E_c / [X_{IIc} E_a + X_{IIa} E_c] \quad (6)$$

$$X_{IIc} + X_{IIa} = 1 \quad y_I + y_{II} = 1 \quad (7)$$

In these formulas, thermal expansion parameters are defined as follows:  $\alpha$ , thermal expansion coefficient of DF;  $\alpha_I$ , thermal expansion coefficient of part I;  $\alpha_{II}$ , thermal expansion coefficient of part II;  $\alpha_c$ , thermal expansion coefficient of crystal part in chain axis;  $\alpha_a$ , thermal expansion coefficient of amorphous region;  $E$ , total tensile modulus of DF in fiber direction;  $E_I$ , tensile modulus of part I;  $E_{II}$ , tensile modulus of part II;  $E_c$ , tensile modulus of crystal in chain direction;  $E_a$ , tensile modulus of amorphous;  $x_{IIc}$ , length of crystal part in part II;  $x_{IIa}$ , length of amorphous part in part II;

$y_I$ , width of part I;  $y_{II}$ , width of part II.  $x_{IIc}$ ,  $x_{IIa}$ ,  $y_I$ , and  $y_{II}$  are the parameters of the mechanical model shown in Figure 6.  $v_a = x_{IIa} y_{II}$  is the volume fraction of the amorphous part.

From the result of X-ray diffraction described in previous section, it is considered that the morphology of crystal and amorphous does not change mostly by  $\gamma$ -ray treatment. Therefore, it is also considered that the parameters of mechanical model,  $x_{IIc}$ ,  $x_{IIa}$ ,  $y_I$ , and  $y_{II}$ , do not change.  $E_c$  is reported as 235–250 GPa,<sup>20,38–40</sup> 315 GPa,<sup>41</sup> and  $E_a$  is reported as 0.3 GPa.<sup>24</sup> The reported values of  $E_c$  and  $E_a$  and the mechanical model shown in Figure 6 suggest that the thermal expansion coefficient of DF in fiber direction is dominated by that in continuous crystal region (part I).<sup>3</sup>

The thermal expansion coefficient of part I ( $\alpha_I$ ), that is to say, that of PE crystal in chain axis ( $\alpha_c$ ) was estimated by X-ray measurements and calculations in previous papers.<sup>20–26</sup> Although the weight-average molecular weight of PE samples used in these works were various values in the range from  $10^4$  to  $10^6$ , the estimated crystal thermal expansion coefficients in chain axis were almost equal to  $-12 \times 10^{-6}/K$  as same as the calculated value.<sup>20–22</sup> Therefore, it is considered that the thermal expansion coefficient of crystal in chain axis ( $\alpha_c$ ) does not depend on the molecular weight, in the range from  $2.6 \times 10^4$  to  $2.0 \times 10^6$ . Furthermore, the reported crystal tensile modulus ( $E_c$ ) does not show the remarkable dependence on molecular weight.<sup>20–22,38–40</sup> Therefore, it is considered that  $\alpha_c$  and  $E_c$  described in the formulas from eqs. (1)–(7) and the mechanical model do not change by decreasing molecular weight with  $\gamma$ -ray treatment.

As aforementioned, it is inferred that the parameters in the mechanical model  $x_{IIc}$ ,  $x_{IIa}$ ,  $y_I$ ,  $y_{II}$ ,  $\alpha_c$ , and  $E_c$  do not change by the  $\gamma$ -ray treatment, in the range of absorbed dose in this experiment. And it is suggested that thermal expansion coefficient in fiber direction does not change in this case and that only the molecular weight changes and the morphology of crystal and amorphous does not change. The direct measurements of tensile modulus ( $E_c$ ) and thermal expansion coefficient ( $\alpha_c$ ) of crystal will be necessary for the certification of aforementioned discussions.

As shown in the result of DSC, the interactions between crystals or between the crystal and amorphous regions are considered to decrease by molecular chain scissions with  $\gamma$ -ray treatment. It is expected that the effect of thermal strain can be observed in the low temperature range under the glass transition temperature ( $T_g$ ; about 170 K). In the low temperature range, the contribution of amorphous to thermal expansion coefficient is expected to increase with increasing the tensile modulus of amorphous region. The measurements of thermal strain in the low temperature range will be necessary for those discussions.

## CONCLUSIONS

Thermal expansion coefficients were measured for high-strength PE fiber (DF) with  $\gamma$ -ray treatment in the temperature range from 213 to 303 K, and the following conclusions were drawn.

1. The molecular weight of DF decreased from  $2 \times 10^6$  to  $3.5 \times 10^4$  by 0.2 MGy absorption and to  $2.6 \times 10^4$  by 0.5 MGy absorption with  $\gamma$ -ray treatment, respectively. This result showed the main chain scission of DF by the  $\gamma$ -ray treatment. The melting peaks of DF observed by DSC became sharper by  $\gamma$ -ray treatment, which suggested the main chain scission in DF by  $\gamma$ -ray treatment.
2. X-ray diffraction of DF did not change by  $\gamma$ -ray treatment, which suggested that the crystal structure of DF did not change by  $\gamma$ -ray treatment.
3. All DFs with and without  $\gamma$ -ray treatment showed negative thermal expansion coefficients.
4. The change of negative thermal expansion of DF by  $\gamma$ -ray treatment was small. It is suggested that negative thermal expansion does not change by only the molecular chain scissions.

As aforementioned, it was suggested that the effect of negative thermal expansion of DF in the temperature range from 213 to 303 K by the molecular chain scissions is small and that the length of extended molecular chains contribute to negative thermal expansion a little.

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