# The Radiation Effect on Thermal Conductivity of High Strength Ultra-High-Molecular-Weight Polyethylene Fiber by γ-Rays

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**ABSTRACT:** To understand the contribution to thermal conductivity by the length of the molecular chains in high strength ultra-high-molecular-weight polyethylene (UHMW-PE) fiber, the thermal conductivity in the range of low temperature was investigated for high-strength UHMW-PE fiber (Toyobo, Dyneema®; hereinafter abbreviated to DF) irradiated by γ-rays (γ-rays treatment) that induce the molecular chains scission. The molecular weight of DF decreased by γ-rays treatment. X-ray diffraction behavior did not change by γ-rays treatment. The melting behavior observed by differential scanning calorimetry showed the main chain scission of DF by γ-rays treatment. Heat capacity decreased slightly by γ-rays treatment. Thermal conductivities of DF with and without γ-rays treatment decreased by decreasing temperature. Thermal conductivity of DF decreased very much by γ-rays treatment. The decrease of thermal conductivity was explained by molecular chain scission in DF by γ-rays treatment and it suggested the possibility of the reduction of mean free path of phonon. The decreasing rates of thermal conductivity by γ-rays treatment were smaller than those of molecular weight. Those differences were explained by the difference of probability of chain scission between the amorphous and crystal region. Those results suggested the contribution of the length of extended molecular chains because of high molecular weight on the thermal conductivity of DF. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 101: 2619–2626, 2006

**Key words:** thermal properties; polyethylene; radiation; fibers; high performance polymers

## INTRODUCTION

Recently, the importance of polymeric materials as electrical insulating or structural materials has been recognized in those areas of cryogenic engineering that require super conducting technology, such as superconducting power equipment, magnetic levitated transportation, medical diagnosis equipment or nuclear fusion reactors, and so on. Their thermal properties, and in particular their coefficient of linear expansion and their thermal conductivity, have been important parameters to apply polymeric materials to these areas. For example, high-strength polyethylene fiber (Toyobo, Dyneema®; hereinafter abbreviated to DF) and composite materials reinforced with DF (DFRP) possess negative coefficient of linear expansion toward the direction of the fiber axis.[1–4] Typical applications for these materials are coil bobbins[5–15] and spacers[9,14,16,17] for super conductors. Those mate-

rials are desired not only as thermal insulators but also as electrical insulating thermal conductors for thermal conductivity.[17,18]

From previous studies concerning thermal properties of polymeric materials, it is well known that the thermal conductivity of amorphous polymers is smaller than that of metals and semiconductors.[19,20] Therefore, these have principally been used as thermal insulators. However, other reports have shown that polymeric crystals possess a high thermal conductivity in the direction in which the molecular chains are covalently bonded, polyethylene crystals being an example.[20–27] Furthermore, there are highly-crystalline and high-strength polymeric fibers that possess thermal conductivity almost as high as metals; these include DF[28–30] and high-strength poly(p-phenylene benzobisoxazole) (PBO) fiber.[28] Practical reports have been published concerning the heat-radiation effects of high-temperature superconducting tape material by high thermal conductivity of DF.[17] It is also reported that DF is used for conduction-cooled superconducting magnetic energy storage (SMES).[18] These indicate that the thermal conductivity of these insulators as well as their negative expansivity are important char-

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acteristics when considering their application in the field of cryogenics.

DF is a high-crystallinity and high strength fiber made from ultra-high-molecular-weight polyethylene (UHMW-PE).[31–34] A previous report elucidated that the thermal conductivity of DF was explained by the fiber structure illustrated in the mechanical serial-parallel model (Takayanagi-model), which consists of a crystalline/amorphous structure.[30] It is considered that the high thermal conductivity of DF is contributed by the thermal conduction along the covalent bonding in the molecular chain direction in continuous crystal region composed of extended molecular chains.[29,30] Eventhough the effect of molecular chain length on thermal conductivity of the UHMW-PE is unknown, the relation between the thermal conductivity and the length of molecular chains of DF is investigated in this work.

It is well known that polyethylene undergoes main chain scission by irradiation with γ-rays in the presence of oxygen.[35,36] In this article, we report the thermal conductivity of DFs affected by main chain scission induced by irradiation with γ-rays for investigation of the relation between the thermal conductivity and the length of main chain to clarify the effect on high thermal conductivity of chain length of DF.

## EXPERIMENTAL

### Sample

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

### Measurements

Molecular weight

To observe the relative decreasing of the length of molecular chains by irradiation with γ-rays, the viscosity-average molecular weight of DFs with and without γ-rays treatment were measured by the viscosity method, using *trans*-decalin solution. The DFs with and without γ-rays treatment were dissolved in decalin with a concentration of 0.0045 g/dL. The relative viscosities at 408 K were measured by using Ubbelhode viscometers in the thermostat. This method was reported elesewhere.[37]

### Structure of crystal and amorphous

X-ray diffraction and melting behavior by differential scanning calorimetry (DSC) were observed to investigate the relative change of crystal structure of DFs by γ-rays treatment. A Rigaku RU-200 (40 kV × 100 mA) was used with X-ray diffraction on Ni-filtered CuKα (λ = 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 weighed at 2 mg, and melting experiments were made in the temperature range from 323 to 523 K at a heating rate of 1°C/min.

### Heat capacity

The heat capacities of DFs were measured by DSC in the temperature range from 210 to 280 K at a heating rate of 10°C/min.

### Thermal conductivity

Thermal conductivity ($\kappa$) was measured by a steady-state heat flow method.[28,29] The measurements of $\kappa$ were carried out on the automated measuring system with thermal controller of a Gifford–MacMahon (GM) cycle helium refrigerator as a cryostat.[38] The fiber samples were prepared by bundling about 6000 monofilaments with length of 25 mm. Both ends of the fiber bundle were fixed by adhesive STYCAST GT. One end of the bundle was attached to the cold stage of a GM refrigerator by mechanical pressing, using indium metal and the adhesive STYCAST GT. A small resistance heater (1 kΩ) was adhered to the other end of the bundle using GE7031varnish. The intermediate positions, where thermocouples were contacted, were bound by fine (0.1 mm) Cu wire. Au (Fe: 0.07 at %)-chromel thermocouples were used as thermometers, which were adhered by GE7031 varnish. The sample space was evacuated to below $10^{-3}$ Pa by an oil diffusion pump for heat insulation. The automated measuring system of $\kappa$ was operated in the temperature range from 10 to 300 K. The $k$ was estimated by the relation $k(mW/cmK) = (Q/\Delta T)(L/S)$, where $Q$ is the heat flow per second, $\Delta T$ the temperature difference between thermometers, $L$ the distance between the thermometers, and $S$ the cross section of the bundle. Details of the determination of $k$ were described elsewhere.[28,29]

## RESULTS AND DISCUSSION

### Molecular weight

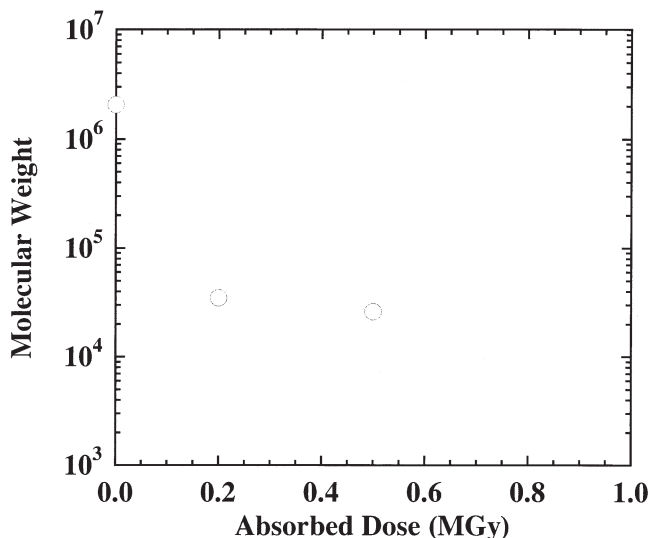The relation between the viscosity-average molecular weight of DF and absorbed dose is shown in Figure 1.

**Figure 1** Dependence of molecular weight of DF on absorbed dose irradiated by γ-rays.



**Figure 2** X-ray diffraction profiles of DFs irradiated and unirradiated with γ-rays.

The measured molecular weight of DF(Blank) was 2.0 × 10⁶. It decreased to 3.5 × 10⁴ by 0.2 MGy irradiation and to 2.6 × 10⁴ by 0.5 MGy irradiation, respectively. This result shows the main chain scission of DF by the γ-rays treatment.

In the irradiation effects of PE in air, the free radicals of alkyl type are produced by irradiation initially, 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 molecular chains. Therefore, the irradiation effects of PE in air depend on the diffusion of oxygen.[31,36] It is considered that oxygen invades and diffuses into the amorphous region more easily than into the crystal region in DF. Therefore, it is 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 γ-rays treatment are investigated. The width, shapes, intensities, and reflection angles of those peaks do not change by γ-rays treatment. It is suggested that the crystal size in (110) and (200) directions do not change. Crystallinities of the DFs cannot be estimated by these results of X-ray diffraction measurements. However, from the independence of X-ray behavior with γ-rays treatment, it is considered that the crystal of DF is destructed by γ-rays not so much. The crosslinking reactions dominat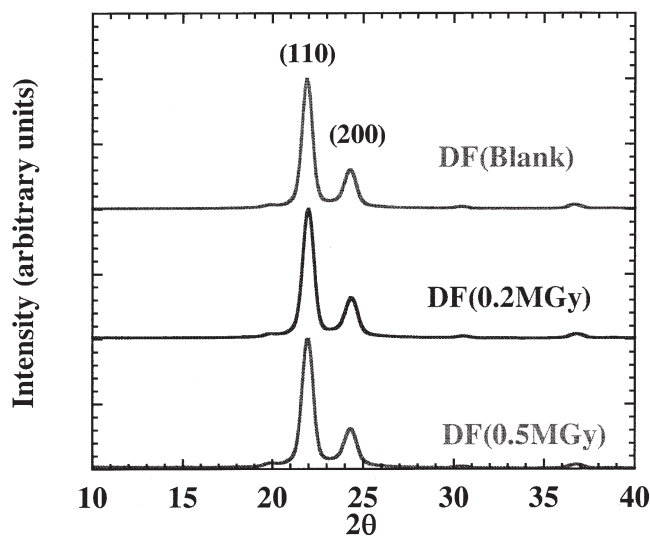e in the case of irradiation of PE in the vacuum. It is reported that X-ray diffraction profile in equatorial direction does not change in the irradiation of γ-rays with absorbed dose of less than 0.25 MGy in vacuum[39] like the case of this work.

**Melting behavior**

The melting behaviors of DFs observed by DSC are shown in Figure 3. DF(Blank) shows four broad melting peaks in the temperature range from 403 to 425 K.
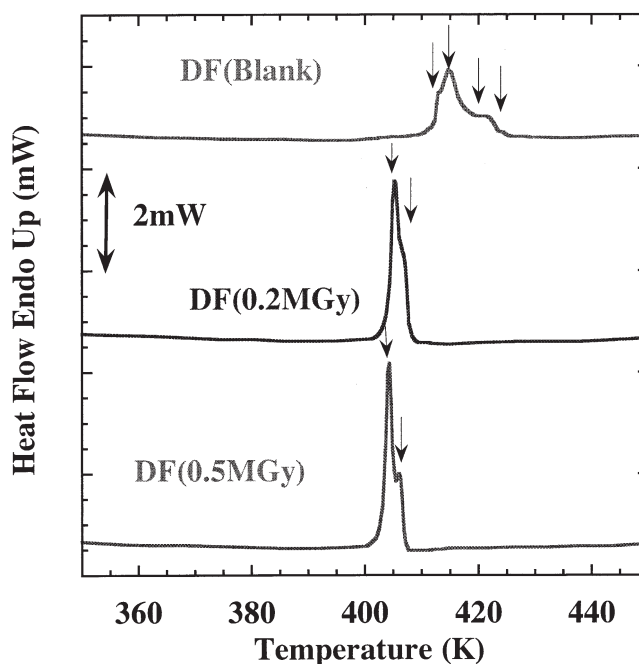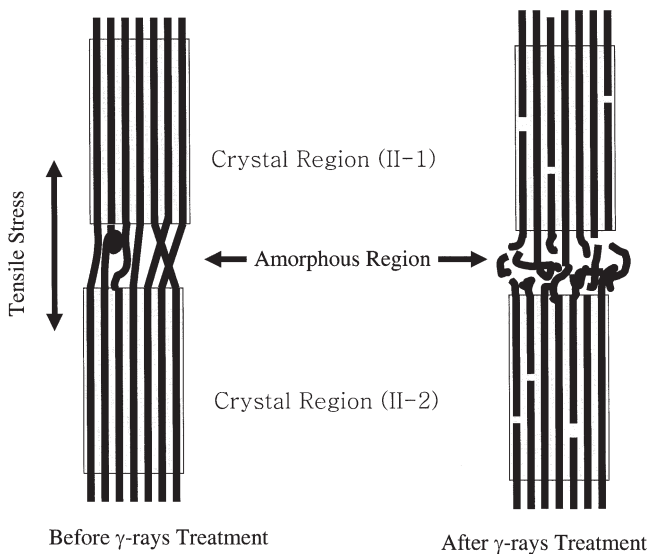


**Figure 3** DSC melting curves of DFs.

**Figure 4** Schematic diagram of molecular chains in series combination part of crystal and amorphous regions of DFs before and after γ-rays treatment.

The melting behaviors of high-strength polyethylene fiber as DF are reported in other articles.[40–42] Although the assignments of the peaks in melting process of high-strength polyethylene fiber are not clarified, it is reported that UHMW-PE molecular chains extended by the tensile stress show the melting behaviors[40–42] like that of DF(Blank). The schematic diagram of the molecular chains in series combination part of crystal and amorphous regions in DF is shown in Figure 4. It is considered that the molecular chains in amorphous of DF(Blank) are extended and are restrained by orientation in the drawing as shown in the left side of Figure 4. 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. Therefore, it is considered that molecular chains in DF(Blank) behave like those in big crystal by the interactions between crystals and that those interactions are not uniformed.

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 4 to 2 after γ-rays treatment. This change suggests the decrease of restraint on molecular chains in crystal region with γ-rays treatment. From the X-ray diffraction studies discussed in the previous section, the crystal size is inferred to decrease not so much by γ-rays treatment. But it is considered that the molecular chains in amorphous region become more movable and that the interactions of molecular chains between the crystal and amorphous or neighboring crystal regions decrease very much by main chain scission

after γ-rays treatment, as shown in the right side of Figure 4. Therefore, it is considered that molecular chains in crystal regions behave more uniformly by decreasing interactions with neighboring amorphous or crystal regions. The sharpe peaks in melting behavior are inferred to be caused by the decrease in those interactions by main chain scissions after γ-rays treatment.

The melting enthalpies ($\Delta H$) and melting temperatures ($T_m$) of DF with and without γ-rays treatment were shown in Figure 5. Both of them decrease by γ-rays treatment though X-ray diffraction behavior does not change. The differences of $\Delta H$ and $T_m$ of DF between with and without γ-rays treatment are remarkable. It is possible for the $\Delta H$ of DF(Blank) to contain the transition energy from orthorhombic to hexagonal, the melting energy of molecular chains in crystal, or restraint of molecular chains in crystal and amorphous-like crystal. Those are considered to be decreased after γ-rays treatment. Especially, the restraint of molecular chains is considered to be decreased by main chain scission. And it is also considered that the molecular chains in the crystal of DF after γ-rays treatment behave as those in small crystal than those in DF(Blank) by decrease of interactions between crystals. Therefore, it is considered that the decreases of interaction between crystal regions by main chain scission induce the decrease of $T_m$. The difference between X-ray diffraction and melting behavior observed by DSC of DFs before and after γ-rays treatment is inferred to be caused by the behavior of restrained molecular chains.

On the other hand, the differences of $\Delta H$ and $T_m$ are small with absorbed dose between 0.2 MGy and 0.5 MGy. It is considered that the decrease of $\Delta H$ and $T_m$ with γ-rays treatment is caused by the decrease of
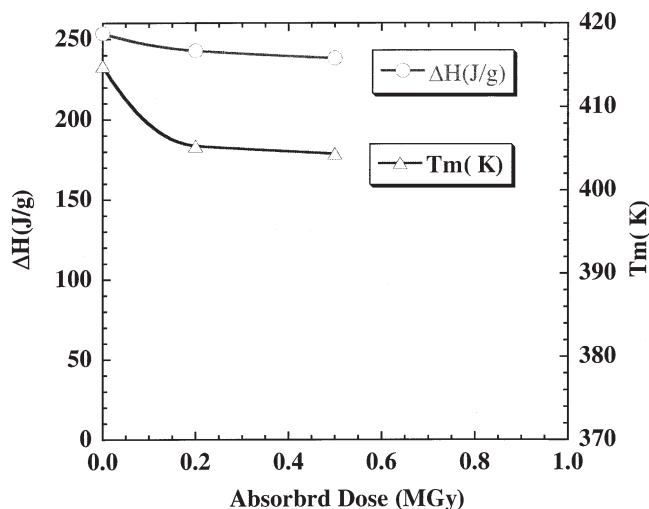


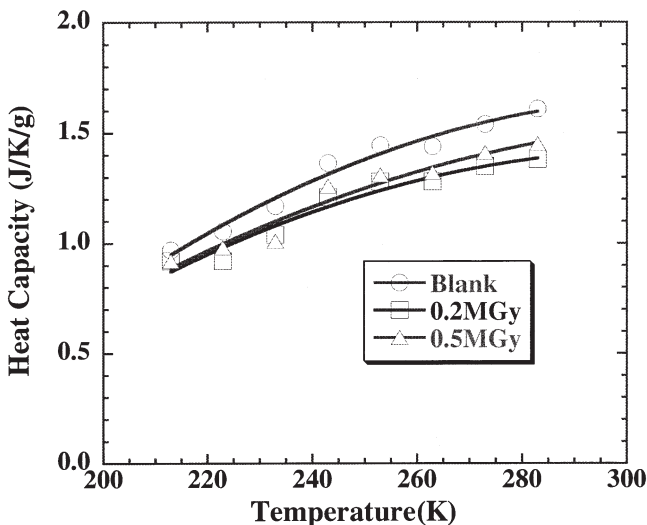**Figure 5** Melting enthalpy ($D\Delta$) and temperature ($T_m$) of DFs.

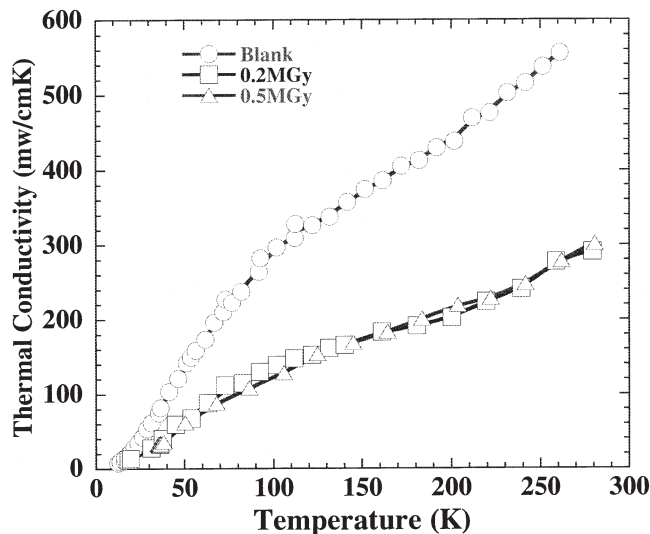**Figure 6** Temperature dependence of heat capacity of DFs with and without γ-rays treatment.



**Figure 7** Temperature dependence of thermal conductivity of DFs with and without γ-rays treatment.

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

With the aforementioned, the melting behavior observed by DSC also suggested the main chain scission in DF by γ-rays treatment.

From the results of molecular weights, X-ray diffraction and melting behavior of DFs before and after γ-rays treatment with absorbed dose of 0.2 MGy and 0.5 MGy, only main chain scissions, could be observed, though the distinct change of crystal structure could not be observed.

### Heat capacity

Heat capacities of DF with and without γ-rays treatment in the temperature range from 210 to 280 K measured by DSC are shown in Figure 6. Heat capacity decreases slightly by γ-rays treatment. The heat capacities of DF (0.2 MGy) and DF (0.5 MGy) show similar behavior. This agrees to the results of melting behavior.

### Thermal conductivity

Temperature dependences of thermal conductivities of DFs in fiber direction from 10 to 260 K are shown in Figure 7. Thermal conductivities of all samples decrease with decreasing temperature.

We investigate the effect of irradiation with γ-rays to thermal conductivity in the following. Thermal conductivity of DF decreases to 50% by γ-rays treatment at every temperature. But the difference of thermal conductivity by absorbed dose between 0.2 MGy and 0.5 MGy is not recognized at every temperature. With

aforementioned, those γ-rays treatments induced the decrease of molecular weight, that is to say, the reduction of length of molecular chain. The relations between the molecular weight and thermal conductivities at 200, 150, 100, and 50 K are shown in Figure 8 for discussion about relation between the molecular chain length and thermal conductivity. The molecular weight of DF (0.2 MGy) and that of DF (0.5 MGy) are not so different. Therefore, it is reasonable that the thermal conductivities of DF (0.2 MGy) and DF (0.5 MGy) are not so different. Figure 8 suggests that the change of thermal conductivity of DF by γ-rays treatment agrees that of molecular chain length. But more detailed study including structural analysis will be necessary for quantitative discussion about the rela-
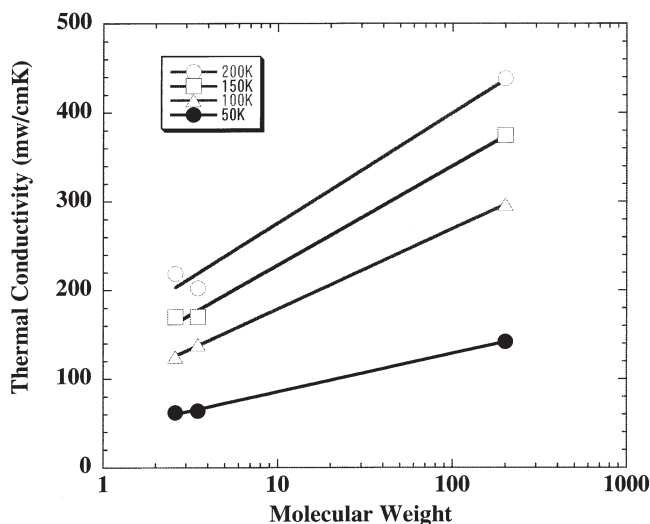


**Figure 8** The relation between thermal conductivity and molecular weight of DFs with γ-rays treatment.

$x_{IIc}$

$x_{IIa}$

$y_I$      $y_{II}$

I            II

Crystal

Amorphous

$x_{IIc}=0.918$
$x_{IIa}=0.082$
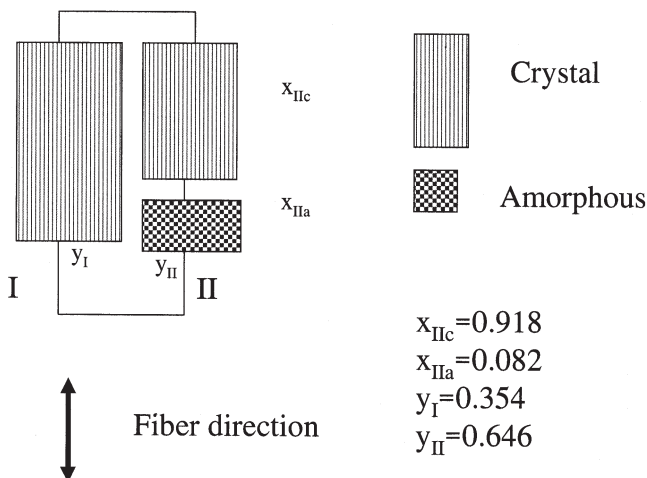$y_I=0.354$
$y_{II}=0.646$

Fiber direction

**Figure 9** Schematic diagram showing the structure of DF by mechanical model.

tion between the absorbed dose and thermal conductivity.

In the next section, the relation between the decrease of thermal conductivity 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 crystal region and amorphous region (Takayanagi Model). In this model, DF is composed of following two parts by parallel combination. One of them is continuous crystal region (hereinafter Part I) and the other is the series combination part of crystal and amorphous region (hereinafter Part II) as shown in Figure 4. In previous articles,[3,30] it is reported that thermal conductivity and thermal expansion coefficient of DF are explained by this serial-parallel mechanical model (Takayanagi Model). The schematic diagram showing the structure of DF by the mechanical model used in the previous articles[3,30] is shown in Figure 9. In the earlier section, only the decrease of molecular weight could be observed as the effect of the γ-rays treatments. The change of crystal structure by γ-rays treatment could not be observed in the X-ray diffraction. Therefore, the effect of irradiation with γ-rays to thermal conductivity is discussed on the assumption that the γ-rays treatments induce only the main chain scission and that the morphology of mechanical model shown in Figure 9 does not change. Thermal conduction of DF in fiber direction is dominated by the thermal conduction in the continuous crystal region (Part I) shown in this figure.[30] The chain axis in the crystal regions is considered as fiber axis to discuss the thermal conductivity of DF in fiber direction.[30] Therefore, the thermal conduction of DF is explained by the thermal conduction along the covalent bonding in the chain axis in the continuous crystal region (Part I).[30] Thermal conductivity is shown in the following formula[1] in the case of phonon conduction.

$$\kappa = (1/3)\, Cp\, \rho\, v\, l \qquad (1)$$

In this formula, the parameters are defined as follows: $\kappa$, thermal conductivity in the fiber direction; $Cp$, heat capacity at constant pressure; $\rho$, density of DF; $v$, sound velocity in fiber direction; $l$, mean free path of phonon in fiber direction. The main chain scission induces the reduction of the length of molecular chain. Therefore, it is inferred that the extended molecular chains composing the crystal region are shortened by the main chain scission with the γ-rays treatment. The reduction of the molecular chain length means the decrease of the average length between the ends of molecular chains. The reduction of the length of molecular chains decreases the mean free path of phonon in Part I in fiber direction, although the morphology of continuous crystal region (Part I) in mechanical model shown in Figure 9 does not change. Furthermore, it is considered that the decrease of mean free path of phonon on Part I decreases the total thermal conductivity of DF.

As described in the earlier section, the heat capacity of DF decreases slightly by γ-rays treatment. On the other hand, the decreases of thermal conductivity by γ-rays treatment are about 50%, and it can not be explained by the slight decrease of heat capacity. This suggests that the decrease of thermal conductivity by about 50% after γ-rays treatment is caused by the decrease of the part except the heat capacity in the formula,[1] that is to say, sound velocity or mean free path of phonon. Those suggest the possibility that the decrease of thermal conductivity is caused by the reduction of mean free path of phonon induced by the main chain scission with γ-rays treatment. The direct measurements of thermal diffusivity and sound velocity will be necessary for certification of this discussion. Furthermore, those results suggest the contribution of high molecular weight on the thermal conductivity of DF.

The relation between the decreasing rate of thermal conductivity and molecular chain length by γ-rays treatment is discussed in the following section. The molecular weight of DF decreases to 1.7% and 1.3% by the γ-rays treatment with the absorbed dose of 0.2 MGy and 0.5 MGy, respectively. It is expected that the average length of molecular chains decrease by the similar rate. In the assumption of the uniformity of the main chain scission in DF, the decreasing rate of thermal conductivity is expected to be the same as that of molecular weight, because it is considered that the reduction of the average length of the molecular chains contribute to the decrease of the mean free path of phonon. But the decreasing rate of thermal conductivity by γ-rays treatment is about 50%. It is considered that the difference of the decreasing rate between the molecular weight and thermal conductivity is due to nonuniformity of main chain scission in DF. The

reason of nonuniformity of the chain scission is discussed by mechanical model composed of crystal and amorphous region in the following section.

It is known that the main chain scission of PE is induced by the oxidation by γ-rays and that it depends on diffusion of oxygen.[36] And it is also known that the diffusion constant of oxygen is larger in amorphous region than in crystal region usually.[36] Therefore, the probability of chain scission is considered to be higher in amorphous region than in crystal region as shown in Figure 4. It is considered that contribution to the thermal conduction of DF by Part II in Figure 9 is small. Therefore, it is considered that the chain scission in amorphous region does not contribute to the decrease of thermal conductivity of DF mostly. The decrease of thermal conductivity is considered to be dominated by the chain scission in crystal region in which the probability of chain scission is less than that in amorphous region. Therefore, the difference of the decreasing rate between thermal conductivity and molecular weight by the γ-rays treatment can be explained by the difference of probability of chain scission between the amorphous and crystal region.

The molecular chains can be oriented by the extension of the molecular chain in DF. The existence of intermediate region between crystal and amorphous region are suggested in some articles.[43–45] The investigation of probability of main chain scission in the crystal and amorphous region will be necessary for quantitative discussion about thermal conductivity of DF with γ-rays treatment.

## CONCLUSIONS

Thermal conductivity was measured for high-strength polyethylene fiber (DF) with γ-rays treatment in the temperature range from 10 to 300 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 γ-rays treatment, respectively. This result showed the main chain scission of DF by the γ-rays treatment. The melting peaks of DF observed by DSC became sharper by γ-rays treatment and that suggested the main chain scission in DF by γ-rays treatment.

2. X-ray diffraction of DF did not change by γ-rays treatment, and the change of the crystal structure of DF by γ-rays treatment could not be observed.

3. Heat capacity decreased slightly by γ-rays treatment.

4. Thermal conductivities of Dfs with and without γ-rays treatment decreased with decreasing temperature.

5. Thermal conductivity of DF decreased to 50% by γ-rays treatment of absorbed dose with 0.2 MGy and 0.5 MGy.

6. The decrease of thermal conductivity of DF by γ-rays treatment was explained by the molecular main chain scission and it suggested the possibility of the reduction of mean free path of phonon.

7. The decreasing rates of thermal conductivity by γ-rays treatment were smaller than those of molecular weight. Those differences were explained by the difference of probability of chain scission between the amorphous and crystal region.

With the aforementioned, thermal conductivity of DF decreased by the main chain scissions with γ-rays treatment. Those results suggested the contribution of the length of extended molecular chains because of high molecular weight on the thermal conductivity of DF.

## References

1. Scholle, K. F. M. J. 9th Int SAMPE European Chapter, Milano, Italy, June 14–16, 1988.
2. Kashima, T.; Yamanaka, A.; Takasugi, S.; Nishihara, S. Adv Cryog Eng 2000, 46, 329.
3. Yamanaka, A.; Kitagawa, T.; Tsutsumi, M.; Kashima, T.; Fujishiro, H.; Ema, K.; Izumi, Y.; Nishijima, S. J Appl Polym Sci 2004, 93, 2918.
4. Kashima, T.; Yamanaka, A.; Nishijima, S.; Okada, T. Adv Cryog Eng 1996, 42, 147.
5. Kamijo, H.; Nemoto, K.; Kashima, T. Meeting on Cryogenics and Superconductivity, 1993, 50, 105.
6. Kashima, T.; Yamanaka, A.; Yoneda, E. S.; Nishijima, S.; Okada, T. Adv Cryog Eng 1996, 41, 441.

7. Yamanaka, A.; Kashima, T.; Nishijima, S.; Okada, T. Cryog Eng 1998, 33, 710.

8. Takao, T.; Watanabe, K.; Kubosaka, T.; Suzuki, T.; Kashima, T.; Yamanaka, A.; Fukui, S. IEEE Trans Appl Supercond 1999, 9, 1133.

9. Takeda, K.; Chiba, M.; Fukuda, K.; Sakagami, Y.; Shibuya, M.; Miyashita, K.; Moriai, H.; Kamata, K. Proc ICEC 1999, 17, 45.

10. Yamanaka, A.; Kashima, T.; Nishijima, S.; Okada, T. Cryog Eng 2000, 35, 530.

11. Yamanaka, A.; Kashima, T.; Hosoyama, K. IEEE Trans Appl Supercond 2001, 11, 4061.

12. Yamanaka, A.; Kashima, T.; Nishijima, S.; Takao, T.; Takeo, M. Cryog Eng 2001, 36, 525.

13. Yamanaka, A.; Kashima, T.; Hosoyama, K.; Nago, S.; Takao, T.; Sato, S.; Takeo, M. Physica C 2002, 372–376, 1447.

14. Takeo, M.; Sato, S.; Takao, T.; Kashima, T.; Yamanaka, A. Cryogenics 2003, 43, 649.

15. Hoshino, T.; Salim, K. M.; Muta, I.; Nakamura, T.; Yamada, M.; Yamanaka, A. Proceedings of Meeting on Cryogenics and Superconductivity 2004, 70, 22.

16. Yamanaka, A.; Kashima, T.; Nishijima, S.; Okada, T. Cryog Eng 1997, 32, 330.

17. Takao, T.; Kawasaki, A.; Yamaguchi, M.; Yamamoto, H.; Niiro, A.; Nakamura, K.; Yamanaka, A. IEEE Trans Appl Superconductivity 2003, 13, 1776.

18. Mito, T.; Kawagoe, A.; Chikaraishi, H.; Okumura, K.; Seo, K.; Maekawa, R.; Henmi, T.; Abe, R.; Baba, T.; Yokota, M.; Morita, Y.; Yamauchi, K.; Hayashi, K.; Iwakuma, M.; Sumiyoshi, F. Proceedings of Meeting on Cryogenics and Superconductivity 2004, 70, 101.

19. Bhowmick, T.; Pattanayak, S. Cryogenics 1990, 30, 116.

20. Jackel, M.; Muller, M.; Claverie, A. L.; Arndt, K. F. Cryogenics 1991, 31 228.

21. Choy, C. L.; Wong, S. P.; Young, K. J Polym Sci Polym Phys Ed 1985, 23, 1495.

22. Choy, C. L.; Leung, W. P. J Polym Sci Polym Phys Ed 1983, 21, 1243.

23. Mergenthaler, D. B.; Pietralla, M.; Roy, S.; Killian, H. G. Macromolecules 1992, 25, 3500.

24. Burgress, S.; Greig, D. J Phys C: Solid State Phys 1975, 8, 1637.

25. Gibson, A. G.; Greig, D.; Sahota, M.; Ward, I. M.; Choy, C. L. J Polym Sci Polym Lett Ed 1977, 15, 183.

26. Choy, C. L. Polymer 1977, 18, 984.

27. Choy, C. L.; Luk, W. H.; Chen, F. C. Polymer 1978, 19, 155.

28. Fujishiro, H.; Ikebe, M.; Kashima, T.; Yamanaka, A. Jpn J Appl Phys 1997, 36, 5633.

29. Fujishiro, H.; Ikebe, M.; Kashima, T.; Yamanaka, A. Jpn J Appl Phys 1998, 37 1994.

30. Yamanaka, A.; Fujishiro, H.; Kashima, T.; Kitagawa, T.; Ema, K.; Izumi, Y.; Ikebe, M.; Nishijima, S. J Polym Sci Part B: Polym Phys Ed, to appear.

31. Izumi, Y.; Niishi, M.; Seguchi, T.; Ema, K.; Yamamoto, T. Radiat Phys Chem 1991, 37, 213.

32. Ohta, Y.; Sugiyama, H.; Yasuda, H. J Polym Sci Part B: Phys Ed 1994, 32, 261.

33. Ohta, Y.; Yasuda, H. Kobunnshi 1995, 44, 658.

34. Ohta, Y. Sen'I Gakkaishi 1998, 54, 8.

35. Ohta, Y. Sen'I Gakkaishi 1999, 55, 413.

36. Ohta, Y. Sen'I Gakkaishi 2004, 60, 451.

37. Fujishiro, H.; Ikebe, M.; Naito, T.; Noto, K. Cryog Eng 1993, 28, 533.

38. Tsubakihara, S.; Yasuniwa, M. Reports on Progress in Polymer Physics in Japan 1993, 36, 187.

39. Xiao, C.; Zhang, Y.; An, S.; Jia, G. J Appl Polym Sci 1996, 59, 931.

40. Fu, Y.; Chen, W.; Pyda, M.; Londono, D.; Annis, B.; Boller, A.; Habenschuss, A.; Cheng, J.; Wunderlich, B. J Macromol Sci Phys B 1996, 35, 37.

41. Shaker, M.; Kamel, I.; Abdel-Bary, E. M. J Elastomers Plast Eng 1996, 28, 236.

42. Kitamaru, R.; Horii, F.; Murayama, M. Macromolecules 1986, 19, 636.

43. Kaji, A.; Ohta, Y.; Yasuda, H.; Murano, M. Polym J 1990, 22, 455.

44. Kaji, A.; Yamanaka, A.; Murano, M. Polym J 1990, 22, 893.

45. Clough, R. L.; Gillen, K. T. In Proceedings of International Symposium on Radiation Degradation of Polymers and the Radiation Resistant materials, Takasaki, Japan, July 24–25, 1989, pp 13–29.