Positron irradiation effects on polypropylene

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Abstract

Positron irradiation effects on polypropylene (PP) and polyethylene during the positron lifetime measurements have been studied in detail. The effect, which was to decrease $I_3$, has been found to be larger in air than in vacuum. For PP, when the lifetime measurements were performed at temperatures above 348K, the radiation effect was scarcely observable. However at low temperature the radiation effect was accumulated and $I_3$ was decreased during the measurements. Irradiation of Co-60 γ-ray up to the dose of 300 MRad also lead to similar decrease in $I_3$, but the decrease due to γ-irradiation was not as large as that due to the positron irradiation. Irradiation effect of positrons appears to be different from that of γ-rays.

1. Introduction

PA is a useful technique to investigate characteristics of materials. However, as reported by Walender et al. /1/, PA must be carefully applied to study polymers. As positron irradiation induces radiation damage to polymers during the measurement of PA, intensities ($I_3$) of long-lived components of o-Ps decrease gradually with the measurement time. They showed that the decrease depends on the kind of polymers. Large decrease was observed for polypropylene (PP), and it was attributed to radicals formed by the positron irradiation. However, it seems that the decrease in $I_3$ is not simply explained by radicals and there are several factors contributing to it /2,3,4/.

2. Experimental

The PA experiments were conducted with a conventional fast-fast coincidence system having a time resolution of 270 ps full width at half
maximum (FWHM). The positron source was prepared by depositing ca. 1 MBq (30,000 μCi) of aqueous $^{22}$NaCl on a Kapton foil of 7 μm thickness and 10x10 mm area. The diameter of the spot of $^{22}$Na source was ca. 2 mm and the dose rate was estimated to be 0.1 Mrad/h at most.

3. Results and discussion

3.1 Irradiation effect on PP and HDPE in air and vacuum

Irradiation effects of positrons on PP and high density polyethylenes (HDPE) were investigated in vacuum and air at 303 K. Intensities of the o-Ps ($I_3$) decreases with elapsed time and the decrease in $I_3$ ($ΔI_3$) can be summarized as follows:

<table>
<thead>
<tr>
<th></th>
<th>PP</th>
<th>HDPE</th>
</tr>
</thead>
<tbody>
<tr>
<td>$ΔI_3$ in 60 h (Vacuo)</td>
<td>-5.7%</td>
<td>-2.8%</td>
</tr>
<tr>
<td>$ΔI_3$ in 60 h (Air)</td>
<td>-7.8%</td>
<td>-3.8%</td>
</tr>
</tbody>
</table>

The decreases in $I_3$ of PP and HDPE is consistent with the previous results, and $ΔI_3$ in air is larger than that in vacuum. The difference between the experiments in air and vacuum is considered to be due to the radiation induced oxidation, which produces >C=O groups in polymers irradiated in air. The carbonyl groups with its large electron affinity may reduce Ps formation in polymers.

Figure 1 shows $I_3$ vs time at 5 different temperatures. Samples were kept for 24h at these temperatures in vacuum and then cooled to 303K.

3.2 The influence at high temperature (Fig.1)

Above 348K, $I_3$ was constant and, as soon as the temperature was restored to 303K, it decreased in the same way as for the unirradiated samples, which indicates irradiation effect is not accumulated at high temperature.

3.3 The influence at low temperature (Fig.2 and Fig.3)

Figure 2 shows $I_3$ vs time for 6 different temperatures: for four...
temperature cases (260K, 240K, 220K, and 195K), the sample temperatures were restored to 303K after 48h and for 100K after 93h. These results indicate that, as soon as the temperatures are restored to 303K, $I_3$ becomes the same value as that of samples irradiated for the same length of time at 303K. At 100K simply $I_3$ increases, whereas above 100K $I_3$ decreases first and then starts to increase. The turning point for increase and the amount of the increase depend on the temperatures. At 100K, the increase in $I_3$ may be explained by the termination of molecular motion/5/, which takes many hours. Also radicals are frozen and hence cross-linkings are suppressed. The cross-linkings occur at room temperature and $I_3$ of these samples increase slowly at 100K. The slow increase is related to relaxation and can be fitted with the so-called Voigt model. Figure 3 shows the fittings.

![Figure 3](image-url)

**Figure 3**, $e^+$ irradiation time effect

![Figure 4](image-url)

**Figure 4**, $\gamma$-irradiation effect on PP

3.4 Comparison with $\gamma$-irradiation (Fig.4)
For $\gamma$-irradiated samples the data were collected within 1h from the start of measurements. For 100 MRad irradiation of $\gamma$-rays, the decrease in $I_3$ was about 3.7% from that of unirradiated samples. However the decrease in $I_3$ for samples irradiated by positron source is about 5.7% after 60h: the dose by the positron irradiation can be estimated to be at most 0.06MRad. This result suggests that the positron irradiation gives larger radiation effect on PP than the $\gamma$-irradiation effect.

References

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/5/ P.Kindl and G.Reiter, phys.stat.sol.(a)104('87)707