Effects of Additives, Photodegradation, and Water-tree Degradation on the Photoluminescence in Polyethylene and Polypropylene

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Photoluminescence (PL) spectra induced by irradiation of ultraviolet photons are compared among low-density polyethylene (LDPE), crosslinked polyethylene (XLPE), and polypropylene (PP). Three PL bands appear around 4.2, 3.6, and 3.1 eV in LDPE and XLPE, while similar three PL bands are observed at similar energies in PP. The PL spectra and their decay profiles are independent of the presence of additives and are also independent of whether the samples were crosslinked or not. These results indicate that neither the additives nor the crosslinking has any significant effects on the respective three PLs in PE and PP. When the sample was pre-irradiated by the ultraviolet photons under different atmospheres (air, O₂, and vacuum), all the PL intensities decrease with the progress of the pre-irradiation regardless of whether the sample is PE or PP. Therefore, all the PLs are considered to result from impurities. In all the pre-irradiated samples, a new PL band appears at 2.9 eV, of which intensity is stronger when the oxygen partial pressure during the pre-irradiation was lower. This PL is considered to be due to photo-induced conjugated double bonds. It has also been confirmed that water-tree degradation in LDPE or in XLPE does not contribute to PL.

Keywords : photoluminescence, polyethylene, polypropylene, additive, photodegradation, water tree

1. Introduction

A considerable amount of research has been done by examining electroluminescence on the degradation processes such as electrical treeing and water treeing which appear in electrically stressed polymers⁽¹⁾⁻⁽⁸⁾. However, electroluminescence in polymers is very complicated and is difficult to elucidate its origin, since it is impossible to control the energy of electrons and is thus impossible to excite any particular luminescence band. It is also very difficult for electroluminescence to measure its lifetime. In order to characterize a luminescence band and to elucidate its origin, measurement of the emission spectrum alone is not enough at all, and observation of its excitation spectrum and evaluation of its lifetime are indispensable. From this viewpoint, a photoluminescence (PL) study using the clearest excitation means, i.e. laser irradiation, must be done prior to studies by electroluminescence. However, only few PL researches have been done for practically important polymeric insulating materials⁽⁹⁾⁻⁽¹²⁾. In the present research, PL induced by ultraviolet (UV) photons in low-density polyethylene (LDPE), crosslinked polyethylene (XLPE), and polypropylene (PP) are examined, focusing the attention especially on the effects of additives, photodegradation, and water-tree degradation.

2. Experimental Procedures

The samples tested are LDPE sheets 500 μm thick, XLPE sheets 500 μm thick, and PP sheets 50 μm thick. These samples, some of which are additive free and the rest contain additives, are named as listed in Table 1. As also shown in Table 1, some samples were pre-irradiated by UV photons from the ArF excimer

laser under different atmospheres (air, O₂, and vacuum) in order to induce photochemical reactions. This pre-irradiation time was set to be 3000 s in the case of LDPE and XLPE and 1000 s in the case of PP. Furthermore, using samples LN and X, effect of water-tree degradation was examined. After the sample surface was scratched by an emery paper, the two samples were sandwiched between an upper water electrode and a lower silver-painted electrode. By applying ac 3.8 kV (root-mean-square value) between the two electrodes for 504 hours, water trees were formed on the surface in contact with the water electrode.

The reason for the different thicknesses being used for PE and PP is that the values of optical absorbance in the UV region become almost equal with these thicknesses. Since the examined polymers are polyolefins, which only contain saturated C-C and C-H bonds that are not luminous, the luminescence contribution of

Table 1. Samples examined

Name	Polymer	Additive	Treatment
LN	LDPE	Free	AR, PI, W
LA	LDPE	AO	AR, PI
X	XLPE	XL	AR, PI, W
PN	PP	Free	AR, PI
PA	PP	AO	AR
PB	PP	N	AR
PC	PP	SA	AR
PD	PP	AO, N, SA	AR

(Note) AO: antioxidant; XL: byproducts of crosslinking agent; N: neutralizer; SA: slipping agent; AR: As-received; PI: Pre-irradiated in air, in O₂, or in vacuum; W: Water-treed.

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chromophores can be easily observed. This is a reason for choosing these polymers for the present research. The polymers were produced by industrial processes and may contain residues of catalysts used for polymerization and inflation. However, any information on this could not be obtained from the suppliers. The sample LA contains 4,4'-thiobis (6-tert-butyl-m-cresol) as an antioxidant, while samples PA, PB, PC, and PD contain tetrakis [methylene-3-(3', 5'-di-*t*-butyl-4'-hydroxyphenyl) propionate] methane as an antioxidant, calcium stearate as a neutralizer, and cis-13-docosenoic acid amide as a slipping agent. The sample X was crosslinked using dicumyl peroxide, and the presence of its byproducts was confirmed by a gas-chromatography/mass-spectrometer⁽¹³⁾.

Using an ArF excimer laser (photon energy: 6.4 eV, pulse width: ca 25 ns, power: ca 150 mJ/pulse, pulse repetition frequency: 1 Hz, Lambda Physik Compex 205) as a photon source, PL spectra were measured in air. Synchrotron radiation (SR) under multi-bunch operation at the BL1B line of UVSOR Facility (Institute for Molecular Science, Okazaki, Japan, beam energy: 750 MeV) was also used to measure PL, PL excitation (PLE), and UV absorption spectra. The PL decay characteristics were measured by a single photon counting method using synchrotron radiation under single-bunch operation. In order to examine the structural changes induced by the UV pre-irradiation or water-tree degradation, the samples were subjected to Fourier-transform infrared absorption (FT-IR) spectroscopy (JEOL JIR-WINSPEC50). All the measurements were done at room temperature.

3. Results and Discussion

The measured PL intensity generally depends on the mutual angles among the photon source, the sample, and the detector. This sometimes makes it difficult to compare the PL intensities among different samples. Therefore, in the following PL figures excepting Fig. 8, the PL intensity will be shown after being normalized by the highest peak height. Furthermore, the PLE spectrum will be shown so that its intensity at the detected photon energy should correspond to the normalized PL intensity.

Figure 1(a) shows the PL spectrum induced in the sample LN by the irradiation of SR photons at 5.6 eV and its three Gaussian-fitted curves. Note that the apparent peak seen around 2.1 eV is not a real PL peak but a stray or fake replica of the 4.2-eV peak. Therefore, the sample LN has three PL component bands around 4.2, 3.6, and 3.1 eV, which are hereafter referred to as A1, A2, and A3, respectively. Figure 1(b) shows the PLE spectra of this PL detected at 4.2, 3.6, and 3.1 eV. Since curves 1 and 2 in Fig. 1(b) can be reasonably assumed to reflect mainly PLs A1 and A2 respectively, their respective PLE peaks are found to be around 5.6 and 5.3 eV. On the other hand, both PLs A2 and A3 contribute to curve 3. By taking this into account, the PLE peak of PL A3 is considered to be around 4.8 eV.

The PL spectrum induced in the sample LA by 5.6-eV photons from SR is shown in Fig. 2(a) together with its Gaussian-fitted sub-components, while Fig. 2(b) shows its PLE spectra detected at each sub-component peak energy at 4.2, 3.6, and 3.1 eV. Furthermore, the PL spectrum with its Gaussian-fitted

Fig. 1. (a) PL spectrum induced by 5.6-eV photons from SR in the sample LN; Curves A1, A2, and A3 represent PLs A1, A2, and A3, respectively, (b) PLE spectra detected at 4.2 eV (curve 1), 3.6 eV (curve 2), and 3.1 eV (curve 3) in the sample LN

Fig. 2. (a) PL spectrum induced by 5.6-eV photons from SR in the sample LA; Curves A1, A2, and A3 represent PLs A1, A2, and A3, respectively, (b) PLE spectra detected at 4.2 eV (curve 1), 3.6 eV (curve 2), and 3.1 eV (curve 3) in the sample LA

Fig. 3. (a) PL spectrum induced by 5.6-eV photons from SR in the sample X; Curves A1, A2, and A3 represent PLs A1, A2, and A3, respectively, (b) PLE spectra detected at 4.2 eV (curve 1), 3.6 eV (curve 2), and 3.1 eV (curve 3) in the sample X

Fig. 4. Decay profiles of PLs A1(a), A2(b), and A3(c) in samples LN (circles), LA (squares), and X (triangles)

Fig. 5. (a) PL spectrum induced by 5.8-eV photons from SR in the sample PN; Curves A1, A2, and A3 represent PLs A1, A2, and A3, respectively, (b) PLE spectra detected at 4.1 eV (curve 1) and 3.6 eV (curve 2) in the sample PN

sub-components similarly induced by 5.6-eV photons in the sample X and its PLE spectra detected at 4.2, 3.6, and 3.1 eV are shown in Figs. 3(a) and 3(b), respectively.

Comparison among Figs. 1(a), 2(a), and 3(a) and that among Figs. 1(b), 2(b), and 3(b) clearly show that the positions of sub-component PL peaks and those of respective PLE peaks are identical among the samples LN, LA, and X. This indicates that the same PLs, A1, A2, and A3 commonly appear in these three samples. The antioxidant, crosslinked structure, or the cross-linking byproducts neither affects the PL spectra nor results in a new PL. In order to confirm this point more clearly, the decay profiles of the PLs were examined for samples LN, LA, and X. Figure 4(a) compares the three decay profiles detected at 4.2 eV,

which shows that they are identical among these three samples. Similar comparisons are shown in Figs. 4(b) and (c) for the PLs detected at 3.6 and 3.1 eV, respectively. A good similarity among the samples is commonly seen for the three PLs. Therefore, it is concluded that good agreements in the three samples are seen throughout the PL spectra, PLE spectra, and decay profiles for the three PLs. This strongly indicates that the same PLs A1, A2, and A3 appear in samples LN, LA, and X. From this, it turns out that neither the antioxidant, the crosslinked structure, nor the crosslinking byproducts contribute to the PL in PE.

Very similar experimental results were observed for PP. As reported in our previous paper⁽¹⁴⁾, the sample PD shows PL around

Fig. 6. Comparison of PL spectra among five PP samples; PL A1(a) and PL A2(b)

Fig. 7. Decay characteristics of PLs A1(a) and A2(b) observed in five PP samples; ○: PN, △: PA, □: PB, ▼: PC, ◆: PD

3.5 to 4.5 eV with its excitation peak from 5.0 to 6.5 eV. Figure 5(a) shows the PL spectrum induced in the sample PN by the irradiation of SR photons at 5.8 eV. The Gaussian-fitted subcomponents shown by dotted curves are in a good agreement with those observed in samples LN, LA, and X shown in Figs. 1(a), 2(a), and 3(a), respectively. Figure 5(b) shows the PLE spectra in the sample PN detected at 4.1 and 3.6 eV. The two PLE spectra are similar in shape and energy to those detected at 4.2 and 3.6 eV in samples LN, LA, and X shown in Figs. 1(b), 2(b), and 3(b), except for the point that the spectral curve 1 detected at 4.1 eV in PN is located at a higher energy. Note that the PLE spectrum of PN detected at 3.1 eV could not be obtained clearly with a good reproducibility since the PL component at 3.1 eV is very small and is superposed with the component at 3.6 eV. Further experiments have shown that all the other PP samples PA, PB, PC, and PD have similar PL and PLE spectra. This indicates that PP also has PL A1, A2, and A3 components, similarly to LDPE and XLPE.

Excepting the small PL component A3, further analyses have been carried out. Figure 6 compares the spectra of PLs A1 and A2 among the five PP samples. Although only the sample PA shows the PL components at slightly different energies, all the other four samples have their PL components A1 and A2 exactly at the respective same energies. Figure 7 shows the decay characteristics of PL components A1 and A2 observed in the five PP samples. The decay of PL A1 is different at its early stage only in the samples PA and PD. This difference could be attributed to a difference in the ratio between the early fast decay and the late slow decay. However, it should be regarded very subtle since the time range of a PL lifetime generally varies very

widely from sub-ps to hours if its mechanism differs. As shown in Fig. 7, all the other decay profiles are identical. Furthermore, comparison between Figs. 4(a) and 7(a) and that between Figs. 4(b) and 7(b) clearly indicate that the PL components A1 and A2 have their respective same lifetimes in LDPE, XLPE, and PP.

In our previous paper⁽¹⁴⁾, we reported that the PLs A1, A2, and A3 in samples LA and PD became weak when the samples had been pre-irradiated by UV photons from the ArF excimer laser. In the same paper⁽¹⁴⁾, we also reported that a new PL peak, referred to as PL B, was induced in these samples around 2.9 eV. As a clearly different point between the PLs A and B, the intensity of PL B increased with the continuous UV-pre-irradiation, while that of PL A decreased. Moreover, the intensity of PL B became larger as the oxygen partial pressure during the pre-irradiation was lower. In order to confirm whether or not similar phenomena are observed, all the other samples were pre-irradiated by 6.4-eV photons from the ArF excimer laser either in vacuum, air, or in oxygen. Figure 8 shows the PL spectra induced by the irradiation of 5.6-eV photons in the pre-irradiated LN, X, and PN samples. Here, all the measurements were done in sequence using the samples mounted on a vertically movable non-rotating rod in order that comparison of the PL intensities among different samples became possible. The PL components A1, A2, and A3 are decreased by the pre-irradiation, and the PL component B with its center around 2.9 eV is induced. The intensity of PL B becomes larger when the oxygen pressure during the pre-irradiation is lower. These effects of pre-irradiation on the PL spectra are quite similar to those observed in the samples LA and PD⁽¹⁴⁾. Note that similar results were observed in all the other samples in both cases that the PL was excited by the 5.6-eV

Fig. 8. Effects of pre-irradiation of UV photons in various atmospheres on PL spectra excited at 5.6 eV in samples LN(a), X(b), and PN(c); Curve 1: as-received, Curve 2: pre-irradiated in air, Curve 3: pre-irradiated in oxygen, Curve 4: pre-irradiated in vacuum

Fig. 9. Decay profiles of PL B in samples LN (circles), X (triangles), and PN (squares)

Fig. 10. Effects of pre-irradiation of UV photons in various atmospheres on FT-IR spectra measured for the sample LA

photons from SR and 6.4-eV photons from the ArF excimer laser. The decay profile of the PL intensity detected at 2.9 eV is compared in Fig. 9 among the pre-irradiated LN, X, and PN samples, which clearly shows that the decay is identical among these three samples. This strongly indicates that the same PL B appears in all the samples by the UV pre-irradiation.

Figure 10 shows effects of pre-irradiation atmosphere on FT-IR spectra measured for the sample LA that had been pre-irradiated by the ArF excimer laser either in oxygen, in air, or in vacuum. The IR absorption due to carbonyl groups appears around 1720 cm^{-1} only when the pre-irradiation was performed in the presence

Fig. 11. Effects of pre-irradiation of UV photons in various atmospheres on UV absorption spectra measured for samples LN(a) and X(b)

of oxygen. Figures 11(a) and (b) respectively show effects of pre-irradiation atmosphere on UV absorption spectra measured for the similarly pre-irradiated LN and X samples. By the pre-irradiation, the UV absorption around 4-5 eV due to polyenes is increased in the order, in oxygen (smallest), in air, and in vacuum (largest) in both samples. If organic polymers are irradiated by ultraviolet photons in vacuum, conjugated carbon-carbon double bonds, namely polyenes will be formed similarly to the case of plasma treatment⁽¹⁵⁾. However, if the irradiation is performed in the presence of oxygen, severe oxidative reactions such as formation of carbonyl groups and scission of bond chains are considered to occur⁽¹⁶⁾. It has been reported that carbonyl groups are not luminous unless a double bond is present next to the carbonyl⁽¹⁷⁾. Namely, only conjugated double bonds can be luminous⁽¹⁵⁾⁽¹⁸⁾. The oxygen-pressure dependence of the 4-5-eV

Fig. 12. PL spectra in the samples LN (solid curve 1) and X (broken curve 2) induced by irradiation of 5.6-eV photons to their water-treed regions

Fig. 13. PLE spectra detected at 4.2 eV (curve 1), 3.6 eV (curve 2), and 3.1 eV (curve 3) in the sample LN with water trees

absorption intensity shown in Fig. 11 is similar to that of the intensity of PL B shown in Fig. 8. Therefore, we can conclude that the conjugated double bonds are responsible for PL B and that the carbonyl groups formed by oxidation do not contribute to the PL B.

At least two candidates are possible as the chromophores for PLs A1, A2, and A3. The first candidate is the α , β -unsaturated carbonyl, which is commonly present in impurities in organic polymers. This carbonyl has the following structure,

Namely, it has a pair of conjugated double bonds and is known to have a PL band around 4 eV⁽¹⁹⁾, at an energy similar to PLs A1, A2, and A3. By the irradiation of UV photons, the α , β -unsaturated carbonyl becomes the β , γ -unsaturated carbonyl that has the following structure,

Namely, the two double bonds in the β , γ -carbonyl are not conjugated, and are not luminous⁽¹⁸⁾. This may explain the disappearance of PLs A1, A2, and A3 in the pre-irradiated polymers. The other candidate is a polynuclear aromatic impurity, which is commonly present in organic polymers, since it is released into the atmosphere in a large quantity during combustion and pyrolysis of fuels and lubricants. Polynuclear aromatics are known to have a PL band around 4 eV^{(10) (11)}, of which intensity decreases if the sample is subjected to irradiation of UV photons⁽¹⁰⁾.

Next, the effect of water-tree degradation is examined. Figure 12 compares PL spectra between the samples LN and X, which were induced by the irradiation of 5.6-eV photons from SR to their water-treed regions. The two spectra are similar in shape. Furthermore, by comparing these spectra with those observed in their respective as-received samples, namely with the spectrum shown in Fig. 1(a) for the sample LN and the one shown in Fig. 3(a) for the sample X, it is clear that there is no appreciable difference between the spectra observed before and after the water-tree degradation. This indicates that the water tree does not contribute to the PLs. In order to clarify this point, their PLE spectra and decay profiles were also examined. Figure 13 shows PLE spectra detected at 4.2, 3.6, and 3.1 eV for the PL observed in the water-treed region of the sample LN. The three PLE spectra agree quite well with those shown in Fig. 1(b). Furthermore, the PL decay was measured at 4.2, 3.6, and 3.1 eV for the PL observed in the sample X with water trees, and Fig. 14 shows the result. The three decay profiles show good agreements with the

Fig. 14. Decay profiles of PLs A1 (circles), A2 (triangles), and A3 (squares) in the sample X with water trees

Fig. 15. Effects of water-tree degradation on FT-IR spectra measured for the sample LN. Solid curve: as-received, broken curve: water-treed

corresponding profiles shown in Fig. 4. These results clearly show that both the PLE spectra and the decay profiles are also identical to those observed in their respective as-received samples. Therefore, it is concluded that the water tree does not contribute to the PLs.

Comparison of FT-IR spectra between the LN samples with and without water trees is shown in Fig. 15. Two sharp peaks due to carboxylate appear around 1540 and 1560 cm^{-1} only in the degraded sample, which agrees with the reports⁽⁵⁾⁽²⁰⁾ that carboxylate is present in water trees. Since carboxylate has no

conjugated double bonds, the present results are consistent with the above-mentioned fact that only conjugated double bonds are luminous.

4. Conclusion

When UV photons are irradiated to LDPE and XLPE, three PL bands appear around 4.2, 3.6, and 3.1 eV, which have excitation peaks around 5.6, 5.3, and 4.8 eV, respectively. These three PL bands appear also in PP. The additives in the polymers do not contribute to the PLs. The three PLs are considered to be due to α,β -unsaturated carbonyls or polynuclear aromatics present in polymers as an impurity, and they gradually decrease with a continuing irradiation of UV photons irrespective of the irradiation atmosphere due to the photo-decomposition of the chromophores. Concomitantly with the disappearance of the three PLs, a new PL band is induced around 2.9 eV. This PL becomes especially intense in the case that the pre-irradiation was done in vacuum, which indicates that conjugated carbon-carbon double bonds are responsible for the PL. By examining PL characteristics in LDPE and XLPE with water trees, it has become clear that the water tree does not contribute to the PLs.

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