Photografting of Methacrylic Acid on Polyethylene Film: Effect of Mixed Solvents Consisting of Water and Organic Solvent

GINTING SUKA IRWAN,[1] SHIN-ICHI KURODA,[1] HITOSHI KUBOTA,[1] TAKASHI KONDO[2]

[1] Department of Chemistry, Faculty of Engineering, Gunma University, Kiryu, Gunma, 376-8515, Japan

[2] Gunma Prefectural Industrial Technology Research Laboratory, Maebashi, Gunma, 371-0845, Japan

ABSTRACT: Photografting of methacrylic acid (MAA) on linear low-density polyethylene film (thickness = 30 μm) was investigated at 60°C in mixed solvents consisting of water and organic solvent with xanthone as the photoiniator. The organic solvents examined were acetone, methanol, tetrahydrofuran, and dioxane. A maximum percentage of grafting was observed at a certain concentration of organic solvent in the mixed solvent, depending on the nature of the organic solvent. Distribution of the grafted chains in the resultant MAA-grafted film, which was examined by scanning electron microscopy and attenuated total-reflection IR spectroscopy, was largely influenced by the use of the mixed solvent. The grafted samples swelled in the alkaline region and shrank in acidic medium, showing a pH-responsive character. The extent of the pH-responsive function was closely related to the location of grafted chains, which could be controlled by proper choice of the composition of organic solvent in the mixed solvent. © 2002 John Wiley & Sons, Inc. J Appl Polym Sci 83: 2454–2461, 2002

Key words: photografting; mixed solvent; linear low-density polyethylene film; methacrylic acid; pH-responsive character; polyethylene; graft copolymers; stimuli-sensitive polymers

INTRODUCTION

The grafting of vinyl monomers onto polyethylene (PE) has been examined in systems initiated by γ rays,[1] plasma,[2] electron beams,[3] and light.[4] The photografting method is relatively simple and practical compared to other initiation methods. Moreover, photografting may cause grafting reactions to occur both on the surface and throughout the entire thickness of the PE substrate, depending on the grafting conditions.[5]

Methacrylic acid (MAA)-grafted polymer films[6–8] exhibit pH-responsive character where the grafted films shrink in acidic media and swell in alkaline media. Kubota and colleagues observed that distribution of grafted chains in the cross-section of the MAA-grafted PE film can be controlled by proper choice of polymerization system,[5,9] either vapor or liquid phases; additives[10]; and polymerization conditions,[11] including monomer concentration and polymerization temperature in the liquid-phase system. Moreover, the functions of grafted PE films, such as wettability,[9,12] moisture permeability,[13] pH-responsive[14]

and temperature-responsive[15] characters, and catalytic activity[16,17] are greatly influenced by the location of grafted chains in the film substrate.

On the other hand, the use of a mixed solvent consisting of water and organic solvent (mixed solvent) has been examined in various grafting systems. In the graftings induced by radiation,[18,19] light,[20–23] and glow discharge[24] and those initiated chemically[25] on various polymer substrates, the use of the organic solvents has been long known to enhance the grafting because the organic solvent component accelerates diffusion of the monomer by swelling the trunk polymer and the grafted chains, where compounds such as methanol (MeOH), dimethyl sulfoxide, dioxane, and methyl ethyl ketone are used as the organic solvent component. It is interesting to study the effect of a mixed solvent on the distribution of grafted chains and the functions of the resulting grafted copolymers.

This article deals with the introduction of MAA-grafted chains into linear low-density polyethylene (LLDPE) film by means of photografting with mixed solvent. The organic solvents employed were chosen for their relatively high polarities and ability to dissolve MAA monomer. In this study, the effects of the use of mixed solvent as grafting conditions on the photografting of MAA on LLDPE film were investigated in terms of grafting behavior, distribution of MAA-grafted chains, and pH-responsive character of the resulting MAA-grafted films.

EXPERIMENTAL

Materials

The PE film used was a LLDPE 30 μm in thickness. The film sample (3 $\times$ 10 cm, 0.07 g) was immersed in an acetone solution containing 0.1 wt % xanthone (XT) and 0.5 wt % poly(vinyl acetate) (weight-average molecular weight = 100,000), removed from the solution, and then dried at room temperature for 2 h under a reduced pressure of 5 torr to give a XT-coated film. The quantity of XT on the film was too small to determine gravimetrically. However, the existence of XT on the film was confirmed by attenuated total-reflectance infrared (ATR-IR) measurement, where a specific absorption band due to the aromatic ring of XT appeared at 1608 cm^{-1}. MAA was purified by distillation under reduced pressure (4 torr at 35°C). XT, hydrogen peroxide (H$_2$O$_2$), and anthra-

quinone-2,7-disulfonate (AQ) were used as the photoinitiator, and hydrophilic organic solvents such as acetone, MeOH, tetrahydrofuran (THF), and dioxane, all of which were reagent grade, were used without further purification.

Photografting

Photografting was carried out in a Pyrex glass tube containing the XT-coated film sample and 30 mL of water or mixed solvent in which a given concentration of MAA (0.39M) was dissolved under nitrogen atmosphere. Irradiation with a high-pressure mercury lamp (400 W) was carried out at 60°C with a Riko rotary photochemical reactor (RH400-10W) (Riko Kagaku Sangyo Co., Ltd., Chiba, Japan), around which the Pyrex glass tubes were rotated. The Pyrex glass tube used in this study transmitted the light with $\lambda > 290$ nm. Polymerized films were extracted for 24 h with hot water to remove the homopolymer of poly-(MAA) and then dried at room temperature for 24 h under a reduced pressure of 5 torr. This extraction[5] is known to be almost sufficient for removal of the homopolymer from the film. Grafted samples with different percentages of graftings in each system were prepared by variation of irradiation time. The percentage of grafting and the percentage of homopolymer were calculated by the following equations:

$$\text{Grafting (\%)} = \frac{\text{Weight of Grafts}}{\text{Weight of Original Film}} \times 100$$

$$\text{Homopolymer (\%)}$$
$$= \frac{\text{Weight of Homopolymer}}{\text{Weight of Original Film}} \times 100$$

Photopolymerization

Photopolymerization was performed in a 3 mm inner diameter. Pyrex glass tube containing an aqueous solution of MAA, in which known concentrations of organic solvent and photoinitiator were dissolved, under nitrogen atmosphere. Irradiation was carried out at 50°C with the same reactor as that described in the previous section. After the polymerization, the solution was poured into a large excess of MeOH to give a product, which was filtered and dried under reduced pressure to a constant weight. Conversion was defined as follows:

Conversion (%)

$$= \frac{\text{Weight of Polymer}}{\text{Weight of Feed Monomer}} \times 100$$

Electron Probe Microanalysis (EPMA)

Carboxylic acid groups in grafted poly(MAA) were converted to the potassium salts by immersion of the grafted film in an aqueous solution of 1.0 wt % potassium hydroxide at 25°C for 24 h and then washed with water. The distribution profile of potassium atoms in the cross-section of the grafted film was measured by an electron probe microanalyzer (model EPM-8000) from Shimazu Co. Ltd. (Kyoto, Japan).

Measurements of ATR-IR Spectra

ATR-IR spectra of the surface layer of grafted film were measured with an IR spectrometer model MAGNA-IR 750 from Nicolet (Madison, WI). The amount of MAA-grafted chains on the film surface layer was expressed in terms of the absorbance ratio of the carbonyl (stretching) band at 1710 cm^{-1} to the methylene (scissoring) band at 1460 cm^{-1}.

Measurements of Dimensional Change

The grafted film (surface area $= S_0$ mm^2) was immersed in pH 4 and pH 8 solutions at 25°C for 24 h, alternately. To adjust the pH of solution, known concentrations of citric acid and dibasic sodium phosphate solution were mixed. After the treatment, excess solution on the film surface was wiped off with a filter paper, and then the surface area (S_1 mm^2) of the treated film was measured. The dimensional change of the grafted film was defined as follows:

$$\text{Dimensional Change (\%)} = \frac{S_1 - S_0}{S_0} \times 100$$

**RESULTS AND DISCUSSION**

Grafting Behavior

The photografting of MAA on LLDPE film in mixed solvent is shown in Figure 1. The organic solvents employed were chosen for their relatively high polarities. The abscissa in the figure represents the quantity of organic solvent in the mixed solvent. The percentage of grafting in the system
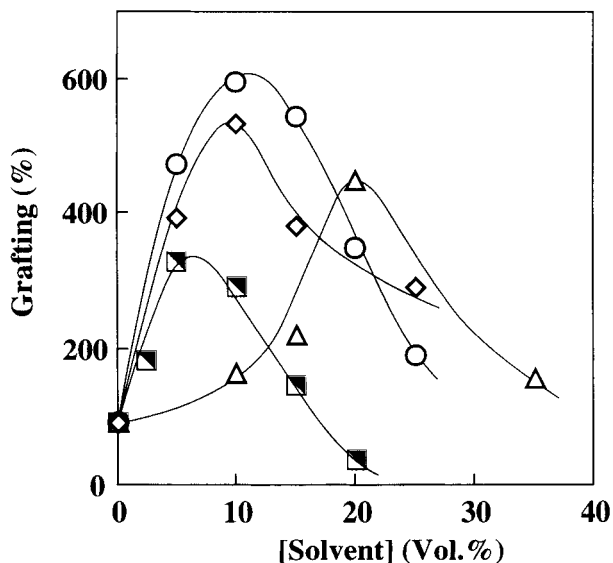
Figure 1 Photografting of MAA on XT-coated LLDPE film in mixed solvent. [MAA] = 0.39 mol/L, [XT] = 0.1 wt %. Irradiation occurred at 60°C for 60 min. (○) acetone, (◇) dioxane, (△) MeOH, and (◣) THF.

without organic solvents was about 90%. When the organic solvents were added to the system, the percentage of grafting increased with increasing concentrations of organic solvent in the mixed solvent, but it decreased beyond a certain concentration of organic solvent, which was commonly observed for each organic solvent employed. Thus, the concentration of organic solvent affording a maximum percentage of grafting depends on the nature of organic solvents. It has been reported that favorable effects of organic solvents have also been found for other polymer substrates besides PE, including styrene–butadiene–styrene triblock copolymer,[19] wool,[25] polypropylene,[26,27] and cellulose,[20] where MeOH, ethanol, dioxane, acetone, and so on were used for the organic solvent component. The percentage of homopolymer, which was expressed as the percentage of the weight of the homopolymer to that of the original film (Fig. 2), decreased with increasing concentrations of organic solvent. This suggests that polymerization of MAA was retarded by the organic solvent in the system.
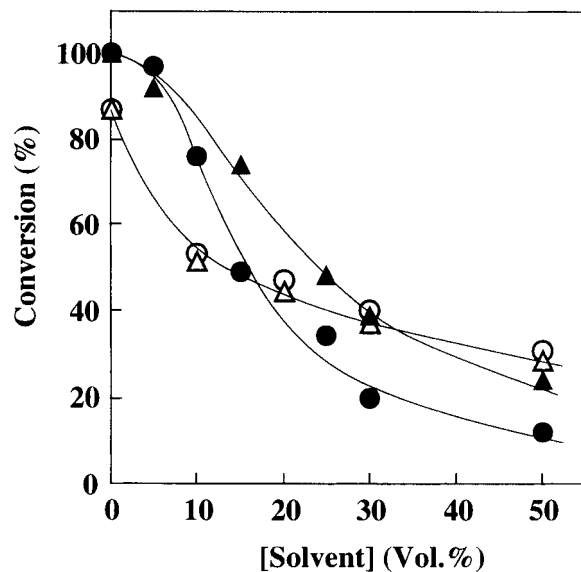
All organic solvents employed were chosen for their relatively high polarities, and the values of the solubility parameter[28] are shown in Table I. It seems that the organic solvents had higher solubility for MAA than water. Therefore, it is reasonable to suppose that monomer solubility in the system increased in the mixed solvent to stimu-
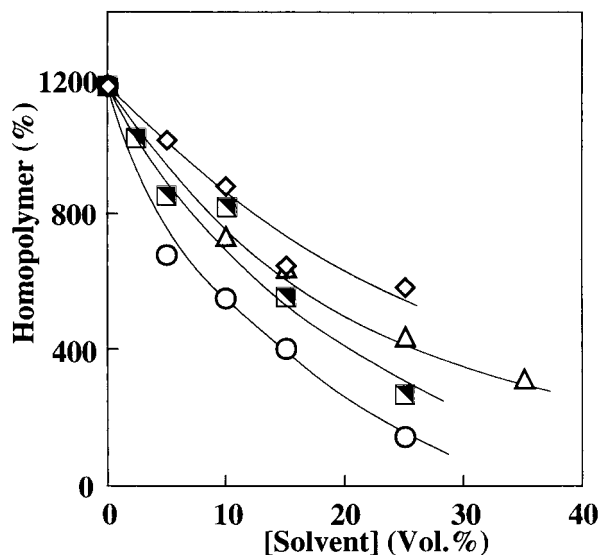
Figure 2 Photografting of MAA on XT-coated LLDPE film in mixed solvent. [MAA] = 0.39 mol/L, [XT] = 0.1 wt %. Irradiation occurred at 60°C for 60 min. (○) acetone, (◇) dioxane, (△) MeOH, and (◼) THF.
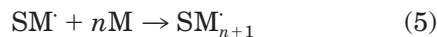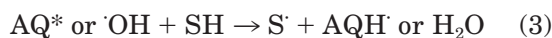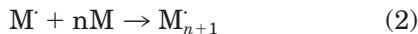

Figure 3 AQ- and H_2O_2-initiated photopolymerizations of MAA in mixed solvent. Irradiation occurred at 50°C for 60 min. [MAA] = 0.39 mol/L, [AQ] = 1 mmol/L: (●) acetone and (▲) MeOH. [H_2O_2] = 1 mmol/L: (○) acetone and (△) MeOH.
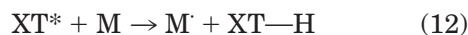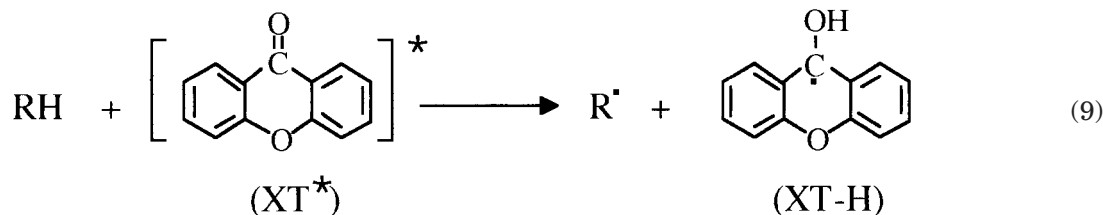
late monomer supply for the radicals on the growing grafted polymer chains and, finally, to increase the percentage of grafting. At a high concentration of organic solvent, on the other hand, the percentage of grafting was supposed to decrease because of the chain transfer from growing polymer radicals to organic solvent and/or the termination of the growing polymer radicals by the solvent radicals formed.

To prove the effect of organic solvent on the photografting of MAA on LLDPE film, we carried out AQ- and H_2O_2-initiated photopolymerizations of MAA in mixed solvent at 50°C for 60 min; the results are shown in Figure 3. AQ[29,30] is known as a water-soluble photoinitiator for the photopolymerization of vinyl monomers. H_2O_2 is known to

Table I   Solubility Parameter[28] of Solvents, Monomer, and Polymer

	Solubility Parameter $(cal/cm^3)^{0.5}$
Water	23.4
MeOH	14.5
Dioxane	10.0
Acetone	9.9
THF	9.1
Poly(MAA)	9.9
MAA	8.5–12.2
PE	8.1

be decomposed by ultraviolet light,[31–33] and the resulting hydroxyl radicals initiate the polymerization of vinyl monomers. As shown in Figure 3, the photopolymerization in the absence of organic solvent (water solvent) proceeded easily, but addition of the organic solvent to the system caused a marked decrease in the conversion with increasing concentrations of organic solvent in the mixed solvent. In the AQ- and H_2O_2-initiated photopolymerizations of MAA in water, the photoexcited AQ molecule (AQ*) and hydroxy radical (˙OH) yielded by photodecomposition of H_2O_2 initiates polymerization according to eqs. (1) and (2), where M and M˙ denote the monomer and its radical, respectively. With the mixed solvent, on the other hand, organic solvent radicals are formed according to eq. (3), where SH and S˙ denote the organic solvent and its radical, respectively. If the solvent radicals have an ability to initiate polymerization, the polymerization will initiate according to eqs. (4) and (5). It is inferred, however, that the possibility of eqs. (4) and (5) is low because the conversion decreases with increasing concentrations of organic solvent in the system, which is shown in Figure 3. Termination occurs mostly by eqs. (6) and (7). Equation (8) is the chain transfer of growing polymer radicals to organic solvent. In a system with a high concentration of organic solvent, eqs. (7) and (8) may be emphasized to result in the decreased conversion:

$$\text{AQ* or } \cdot\text{OH} + \text{M} \rightarrow \text{M}\cdot \text{ or HOM}\cdot \tag{1}$$

$$\text{M}\cdot + n\text{M} \rightarrow \text{M}\cdot_{n+1} \tag{2}$$

$$\text{AQ* or } \cdot\text{OH} + \text{SH} \rightarrow \text{S}\cdot + \text{AQH}\cdot \text{ or H}_2\text{O} \tag{3}$$

$$\text{S}\cdot + \text{M} \rightarrow \text{SM}\cdot \tag{4}$$

$$\text{SM}\cdot + n\text{M} \rightarrow \text{SM}\cdot_{n+1} \tag{5}$$

$$2\text{M}\cdot_{n+1} \rightarrow \text{Polymer} \tag{6}$$

$$\text{M}\cdot_{n+1} + \text{S}\cdot \rightarrow \text{Polymer} \tag{7}$$

$$\text{M}\cdot_{n+1} + \text{SH} \rightarrow \text{Polymer} + \text{S}\cdot \tag{8}$$

Based on the previous investigations, the photografting of MAA on LLDPE film is conceivable to proceed as follows:

$$\text{RH} + [\text{XT}^*] \rightarrow \text{R}\cdot + (\text{XT-H}) \tag{9}$$

$$\text{R}\cdot + \text{M} \rightarrow \text{R---M}\cdot \tag{10}$$

$$\text{R---M}\cdot + \text{M} \rightarrow \text{G}\cdot \tag{11}$$

$$\text{XT*} + \text{M} \rightarrow \text{M}\cdot + \text{XT---H} \tag{12}$$

$$\text{M}\cdot + \text{M} \rightarrow \text{H}\cdot \tag{13}$$

$$2\text{G}\cdot \rightarrow \text{Grafted Polymer} \tag{14}$$

$$2\text{H}\cdot \rightarrow \text{Homopolymer} \tag{15}$$

$$\text{SM}\cdot + n\text{M} \rightarrow \text{H}\cdot \tag{18}$$

$$\text{G}\cdot + \text{S}\cdot \rightarrow \text{Grafted Polymer} \tag{19}$$

$$\text{G}\cdot + \text{SH} \rightarrow \text{Grafted Polymer} + \text{S}\cdot \tag{20}$$

$$\text{H}\cdot + \text{S}\cdot \rightarrow \text{Homopolymer} \tag{21}$$

$$\text{S}\cdot + \text{SH} \rightarrow \text{Homopolymer} + \text{S}\cdot \tag{22}$$

Photografting in water solvent proceeds according to eqs. (9–15), where RH, R·, G·, and H· denote LLDPE and its radicals, and the growing grafted polymer and homopolymer radicals, respectively. Equation (9) is the formation of LLDPE radicals due to the hydrogen-abstracting reaction by photoexcited XT. Equations (10) and (11) lead to the formation of the grafted polymer, while eqs. (12) and (13) are the formation of the homopolymer. Terminations of the growing grafted polymer and homopolymer radicals mostly occur by eqs. (14) and (15), respectively. With the photografting in the mixed solvent system, eqs. (16–22) may proceed in addition to eqs. (9–15) in the photografting in water solvent.

$$\text{XT*} + \text{SH} \rightarrow \text{S}\cdot + \text{XT---H} \tag{16}$$

$$\text{S}\cdot + \text{M} \rightarrow \text{SM}\cdot \tag{17}$$
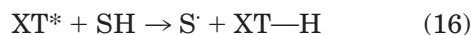
It seems that the increased solubility of MAA monomer due to the organic solvent leads to an emphasized reaction of eqs. (10) and (11), resulting in the increased percentage of grafting. The solvent radicals formed by eq. (16) may participate in the termination of the growing grafted polymer and homopolymer radicals according to eqs. (19) and (21), respectively. Equations (20) and (22) are the chain transfer of grafted polymer and homopolymer radicals to the organic solvent component, respectively. At a high concentration of organic solvent, the termination by the solvent radicals and/or the chain transfer to the organic solvents may be emphasized to result in the reduced percentage of grafting and percentage of homopolymer as shown in Figures 1 and 2.
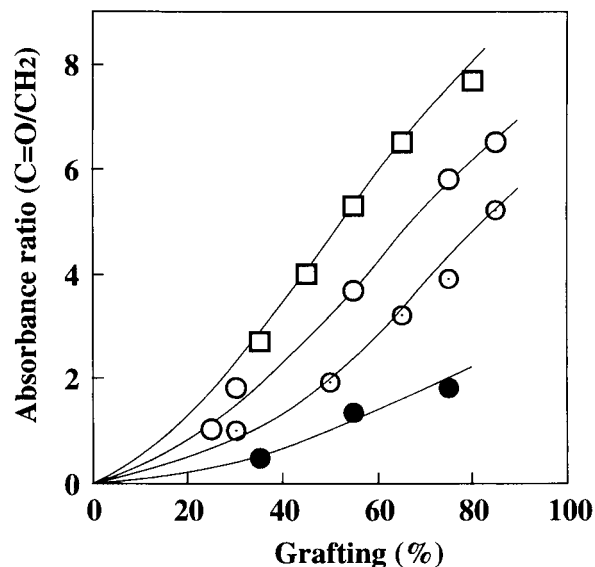
Distribution of MAA-Grafted Chains

Figure 4 shows the absorbance ratio of the carbonyl peak at 1710 cm^{-1} to the methylene peak at 1460 cm^{-1} of the grafted films (sample with mixed solvent) prepared in the mixed solvent. The
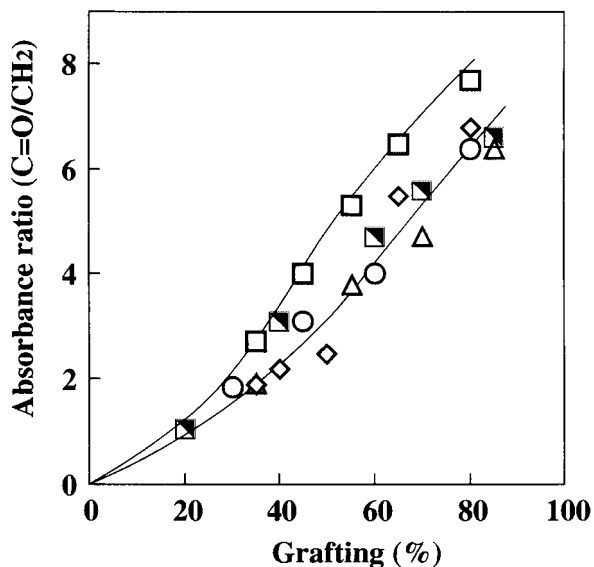
Figure 4 Changes in the absorbance ratio of a carbonyl peak at 1710 cm^{-1} to a methylene peak at 1460 cm^{-1} with percentage of grafting in MAA-grafted LLDPE films: (□) water, (○) acetone (5 vol %), (◇) dioxane (5 vol %), (△) MeOH (5 vol %) and (◣) THF (5 vol %).

Figure 6 Changes in the absorbance ratio of a carbonyl peak at 1710 cm^{-1} to a methylene peak at 1460 cm^{-1} with percentage of grafting in MAA-grafted LLDPE films: (□) water, (○) acetone (5 vol %), (◉) acetone (10 vol %), and (●) acetone (15 vol %).

absorbance ratio increased with an increase in the percentage of grafting, and the magnitude of the increase was slightly lower for the sample with mixed solvent than the sample prepared with water solvent. There was no large difference in the absorbance ratios among the samples with mixed solvent. Figure 5 shows the distribution profile of potassium atoms in the cross-section of MAA-grafted LLDPE films. The grafted chains did not penetrate into the center parts of the film, which was commonly observed with each grafted sample. The effect of the concentration of organic solvent, such as acetone, in the mixed solvent on

the absorbance ratio of the carbonyl peak to the methylene peak was examined, and the results are shown in Figure 6. The absorbance ratio decreased in the order water > 5 > 10 > 15 vol % acetone. The density of the MAA-grafted chains in the surface layer of the grafted film was found to decrease with increasing concentrations of acetone in the mixed solvent. As seen from the results of EPMA measurement (Fig. 7), MAA-grafted chains had a tendency to penetrate into the inside of the film, depending on the concentration of acetone in the mixed solvent. That is, a higher acetone concentration in the mixed solvent resulted in a grafted film with a homogeneous
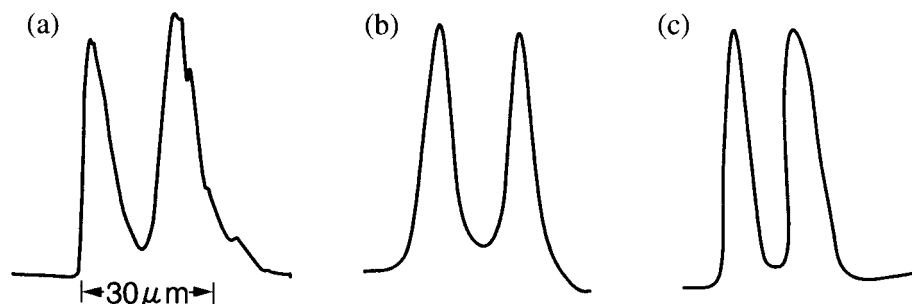
Figure 5 Distribution profile of potassium atoms in the cross-section of MAA-grafted LLDPE films prepared in mixed solvent and measured by EPMA: (a) water (grafting = 65%), (b) acetone (5 vol %; grafting = 58%), and (c) MeOH (5 vol %; grafting = 60%).
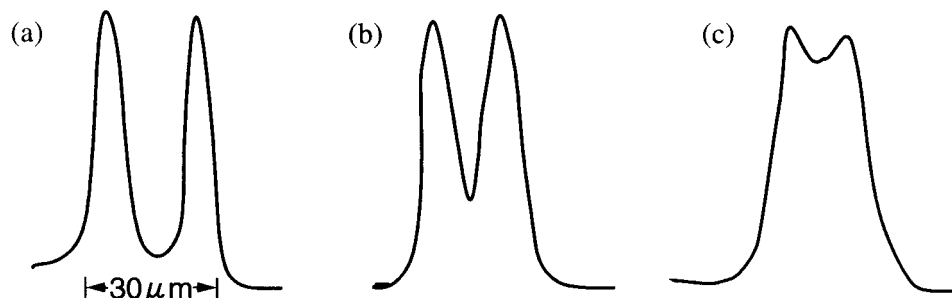
Figure 7 Distribution profile of potassium atoms in the cross-section of MAA-grafted LLDPE films prepared in mixed solvent and measured by EPMA: (a) acetone (5 vol %; grafting = 60%), (b) acetone (10 vol %; grafting = 61%), and (c) acetone (15 vol %; grafting = 62%).

distribution of grafted chains. As indicated from the previous discussion that all organic solvents used are able to dissolve MAA monomer, the monomer is supplied to the LLDPE substrate through the grafted layer, resulting in the penetration of the grafted layer into the LLDPE film texture. Because affinity to the LLDPE substrate is higher for acetone than water, based on the solubility parameters in Table I, moreover, acetone molecules assist the penetration of MAA monomer into the inside of the film, resulting in the penetration of MAA-grafted chains into inside of the film. As noted previously, organic solvents in the mixed solvent influenced both the grafting behaviors and the location of MAA-grafted chains in the LLDPE film substrate.
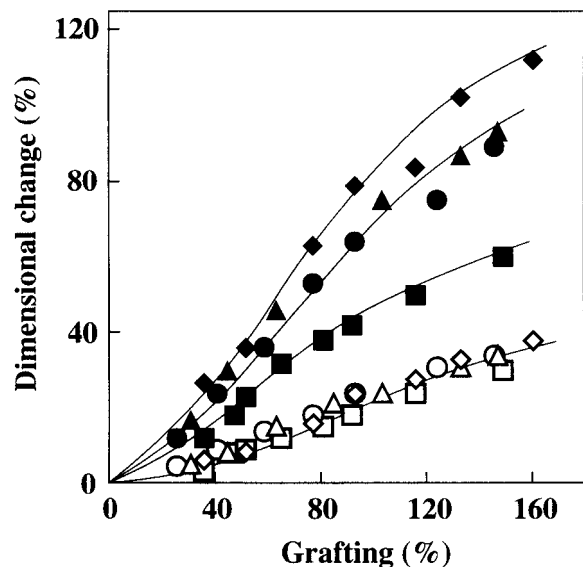
pH-Responsive Characters

It is known that MAA-grafted polymer films exhibit pH-responsive characters, where they shrink in acidic media and swell in alkaline media. This behavior[34] is due to electrostatic repulsions between the carboxylic acids and ions present in the buffered solutions. At high pH values, the pendant carboxylate side chains are repelled by the anions in the solution and expand to minimize charge concentration. Figure 8 shows the dimensional change of the sample with mixed solvent. The dimensional change increased with an increase in the percentage of grafting, and there was no large difference in the dimensional change at pH 4 among various samples, but the magnitude at pH 8 was larger for the sample with mixed solvents than the sample with water. The effect of acetone concentration in the mixed solvent on the dimensional change is shown in Figure 9. The dimensional change increased with the percentage of grafting, and the magnitude decreased in the order 15 > 10 > 5 vol % > water. This might be ascribed to the grafted chains of the sample with mixed solvent located in the film inside compared to the sample with water, which was confirmed by ATR-IR and EPMA measurements (Figs. 6 and 7). It was thus observed that MAA-grafted LLDPE films exhibited a pH-responsive character, which was largely influenced by location of the grafted chain in the grafted film. That is, the grafted film with the grafted chains located inward in the film exhibited a larger ex-
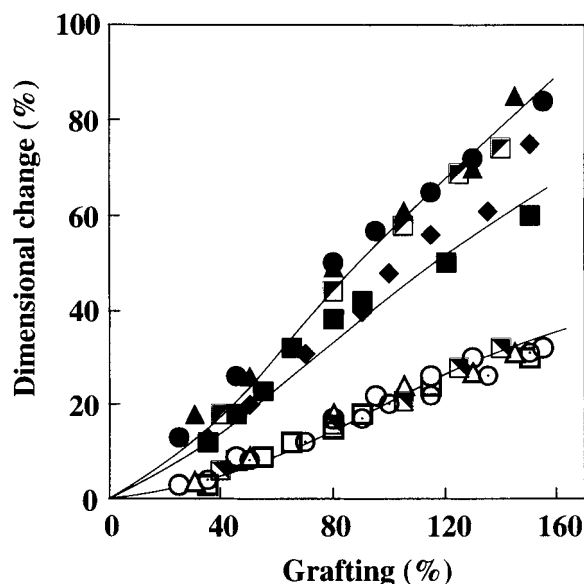
Figure 8 pH-Responsive character of MAA-grafted LLDPE films prepared in water and mixed solvent. [MAA] = 0.39 mol/L. pH 4: (□) water, (○) acetone (5 vol %), (◇) dioxane (5 vol %), (△) MeOH (5 vol %), and (◨) THF (5 vol %). pH 8: (■) water, (●) acetone (5 vol %), (◆) dioxane (5 vol %), (▲) MeOH (5 vol %), and (◪) THF (5 vol %).

Figure 9 pH-Responsive character of MAA-grafted LLDPE films prepared in water and mixed solvent. [MAA] = 0.39 mol/L. pH 4: (□) water, (○) acetone (5 vol %), (△) acetone (10 vol %), and (◇) acetone (15 vol %). pH 8: (■) water, (●) acetone (5 vol %), (▲) acetone (10 vol %), and (◆) acetone (15 vol %).

tent of the pH-responsive character compared to that with the grafted chains located mainly at the surface of the film.

CONCLUSIONS

Based on the previous investigations, it is concluded that MAA-grafted chains having a pH-responsive function can be introduced into LLDPE film by photografting with XT-coated film. The pH-responsive function of the resulting MAA-grafted film is largely influenced by the use of mixed solvent consisting of water and organic solvent. The extent of the pH-responsive function is closely related to the location of grafted chains, which can be controlled by proper choice of the concentration of organic solvent in the mixed solvent.

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