

# Water Crosslinking Reactions of Silane-Grafted Polyolefin Blends

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**ABSTRACT:** Water crosslinking reactions of LDPE, PP, LLDPE, and the LDPE/PP and LDPE/LLDPE blends are investigated. Degrees of crosslinking of these samples are quantitatively compared and discussed in detail in terms of crosslinking ability, phase-separation behavior, molecular weight (or viscosity), morphology of the constituents in blends, or in the pure state. It is found that PP gives negligible crosslinking reactions in the pure state and in blends with LDPE. LDPE and LLDPE are both capable of giving considerable degrees of crosslinking, with LLDPE giving a higher degree of crosslinking than LDPE at all conditions studied. Degrees of crosslinking of the LDPE/LLDPE blends are not linearly but are zigzagly related to the compositions of the blends with the LDPE/LLDPE = 50/50 blend giving a relatively high degree of crosslinking at 100, 120, and 140°C for a certain time except the condition at 140°C for 12 h compared with the LDPE/LLDPE = 75/25 and 25/75 blends. © 2001 John Wiley & Sons, Inc. *J Appl Polym Sci* 81: 1799–1807, 2001

**Key words:** LDPE; LLDPE; PP; grafting reactions; crosslinking reactions

## INTRODUCTION

Crosslinking of polyethylene is of considerable practical importance. It extends the range of use by raising the upper temperature limit of application, and improves the mechanical properties of this polymer. There are a variety of ways to achieve crosslinking of polyethylene.<sup>1</sup> Polyethylene has no functional groups that can provide crosslinking capability, such as the kind one finds in typical thermosetting resins. The most common way in crosslinking polyethylene is through the use of peroxide crosslinking agents added to the resins. Peroxides provide a source of free rad-

icals when heated above their decomposition temperatures. These free radicals are capable of extracting a hydrogen from the polyethylene backbone, thus transferring the free radical site to the polyethylene. With this accomplished, two polyethylene chains can crosslink. A second method of crosslinking polyethylene is through the use of irradiation. In this case, the free radical formed on the polyethylene backbone is the result of electron beam irradiation. The third type of crosslinking is through the grafting of a silane onto the polyethylene and subsequently through the condensation reaction of the silane graft by moisture. The way of making crosslinkable polyethylene through silane grafting followed by water crosslinking<sup>1</sup> has gained much attention in recent years because of its various advantages, such as easy processing, low cost and capital investment, and favorable properties in the processed materials.

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The most common silane used in the manufacture of silane crosslinkable polyethylene is vinyl trimethoxysilane. This silane has conventionally been introduced into polyethylene by melt grafting using a peroxide. In producing the crosslinkable silane-grafted polyethylene, a peroxide is mixed with the silane and polyethylene, and all these components are compounded in an extruder at high temperatures. The silane-grafted polyethylene is then crosslinked through hydrolysis of the silyl trimethoxy groups with water, followed by condensation of the formed silanol groups.<sup>2</sup>

Silane grafting and water crosslinking reactions of LDPE and relevant changes in thermal properties and physical properties of the polymer due to these reactions have been reported.<sup>3-12</sup> In our previous report,<sup>12</sup> the extents of grafting reactions of three polyolefins, including LDPE, HDPE, and LLDPE, were quantitatively compared and found in the order of LLDPE > LDPE > HDPE. It was suggested that LLDPE or LDPE could give higher extent of crosslinking reactions than HDPE. The LLDPE studied<sup>12</sup> was a copolymer of ethylene and 3.5 mol % of 1-hexene comonomer, and has a melting point at around 125°C. The HDPE studied has a melting point at around 138°C. By reacting with water at 100°C, these three silane-grafted polyolefins do not guarantee the extents of crosslinking reactions being in the same order as those of the grafting reactions because considerable diffusion control of water may be developed in LLDPE and HDPE at 100°C of reaction temperature. In addition, polypropylene (PP), also a polyolefin, has not been reported in literature on its silane grafting and water crosslinking reactions. It is thus proposed that silane grafting reactions of PP and extents of crosslinking reactions of PP and two previously studied silane-grafted LDPE and LLDPE be investigated and quantitatively compared. Second, to further increase the upper temperature limit of application for LDPE, it is worthwhile to study the crosslinking reactions of the blends of LDPE with another polyolefin having a higher melting temperature than LDPE, such as PP or LLDPE (having a low comonomer content).

In this article, water crosslinking reactions of LDPE, PP, LLDPE, and the binary blends of these three polyolefins were studied. Factors that influence the extents of the reactions were examined. These factors include temperature, time, composition, and morphology of the blend. Fourier transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC), and polar-

ized optical microscopy (POM) are used for this study.

## EXPERIMENTAL

### Materials

All materials were used as received. LDPE (LE4074) was received from Borealis Corporation in Portugal with a melt index of 2.8 g/10 min and a density of 0.917 g/cm<sup>3</sup>. LLDPE (AL3108ZD) was received from Exxon Corporation with 3.5 mol % of 1-hexene comonomer in the copolymer, a melt index of 0.8 g/10 min, and a density of 0.921 g/cm<sup>3</sup>. PP (PC366) was obtained from the Taiwan Polypropylene Company with a melt index of 1.5 g/10 min. Melt indices were determined according to ASTM D1238 at 190°C and 2.16 kg of loading for all three polymers. Dicumyl peroxide was used as an initiator for grafting reactions and supplied by Aldrich Chemical Company (Milwaukee, WI). Vinyl trimethoxysilane was received from Tokyo Kasei Kogyo Co (Tokyo, Japan).

### Silane Grafting Reactions

Silane-grafted LDPE, LLDPE, and PP were individually prepared by a melt extrusion of the polymer with 0.05 phr (part of reagent per hundred parts of the polymer) of dicumyl peroxide and 5 phr of vinyl trimethoxysilane. A single screw extruder was used for the silane grafting reactions, and was operated at a speed of 10 rpm and at temperatures ranging from 180°C at the feeding zone to 200°C at the compression zone to 160°C at the metering zone of the extruder (30 mm in diameter, L/D, 19, compression ratio, 3.5).

### Preparation of Polyolefin Blends

Melt blending was carried out using the single-screw extruder. For LDPE/PP blends, the extruder was operated at 170°C at the feeding zone, 190°C at the compression zone, and 170°C at the metering zone. For LDPE/LLDPE blends, the extruder was operated at 150°C at the feeding zone, 160°C at the compression zone, and 150°C at the metering zone. The components in a binary blend include both before and after silane grafting reactions. Thus, LDPE/LLDPE = 50/50 was the blend of a mixture of 50 parts by weight of LDPE and 50 parts by weight of LLDPE, and S-LDPE/S-LLDPE = 75/25 was the blend of a mixture of 75 parts by weight of silane-grafted LDPE and 25

**Table I** Descriptions for Some Representative Compositions of Blends Containing LDPE and LLDPE before and after Silane Grafting Reactions, Water Crosslinking Reactions, and Decalin Extractions

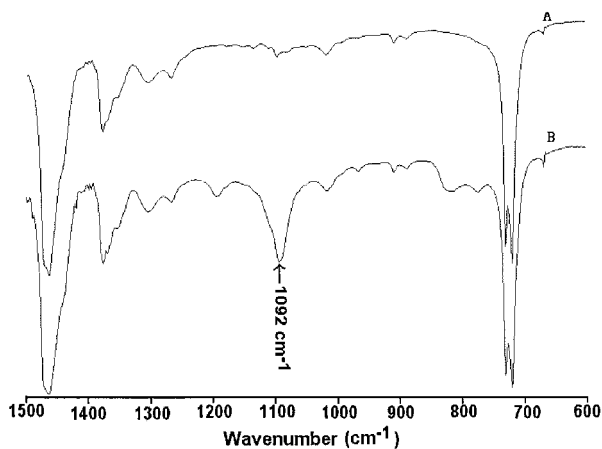
Blend Compositions	Descriptions
LDPE/LLDPE = 100/0	Pure LDPE
LDPE/LLDPE = m/n	The blend of a mixture of m parts by weight of LDPE and n parts by weight of LLDPE
LDPE/LLDPE = 0/100	Pure LLDPE
S-LDPE/S-LLDPE = 100/0	Pure silane-grafted LDPE
S-LDPE/S-LLDPE = m/n	The blend of a mixture of m parts by weight of silane-grafted LDPE (S-LDPE) and n parts by weight of silane-grafted LLDPE (S-LLDPE)
S-LDPE/S-LLDPE = 0/100	Pure silane-grafted LLDPE
X-LDPE/X-LLDPE = 100/0	The water-crosslinked S-LDPE
X-LDPE/X-LLDPE = m/n	The water-crosslinked S-LDPE/S-LLDPE = m/n blend
X-LDPE/X-LLDPE = 0/100	The water-crosslinked S-LLDPE
E-LDPE/E-LLDPE = 100/0	The remainder after decalin extraction of X-LDPE
E-LDPE/E-LLDPE = m/n	The blend remainder after decalin extraction of the X-LDPE/X-LLDPE = m/n blend
E-LDPE/E-LLDPE = 0/100	The remainder after decalin extraction of X-LLDPE

parts by weight of silane-grafted LLDPE, to give two examples. The LDPE/LLDPE (or LDPE/PP) blends were then compressed in a hot press at 150°C (or 180°C for LDPE/PP blends) for 30 s to make film samples with around 0.45 mm in thickness for water crosslinking reactions.

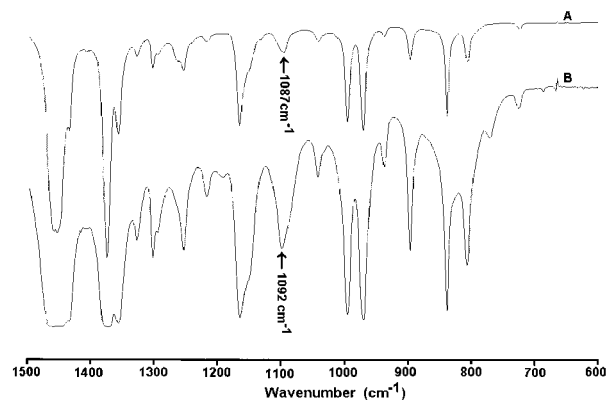
### Water Crosslinking Reactions

The silane-grafted film samples were immersed into water at a controlled temperature for a certain time to conduct crosslinking reactions. Degrees of crosslinking of the water-crosslinked samples were measured by determining their gel

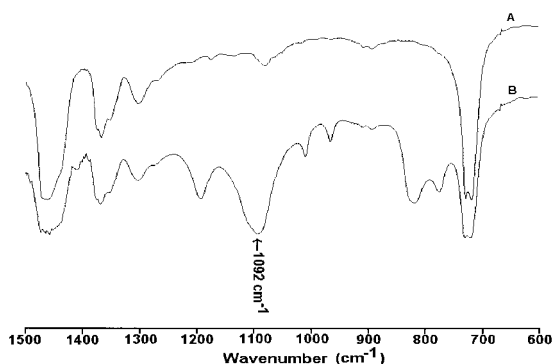
contents after extraction of the soluble component from the samples in decalin refluxing for 6 h. X-LDPE/X-LLDPE = 25/75 denotes the water-crosslinked product from the S-LDPE/S-LLDPE = 25/75 blend. E-LDPE/E-LLDPE = 50/50 denotes the remainder after decalin extraction of the X-LDPE/X-LLDPE = 50/50 blend. Table I lists descriptions for some representative compositions of blends containing various polyolefins before and after silane grafting reactions (with a prefix S for the polyolefin), water crosslinking reactions (with a prefix X for the polyolefin), and decalin extractions (with a prefix E for the polyolefin).



**Figure 1** FTIR spectra of LDPE (A) before and (B) after grafting reactions with 5 phr of vinyl trimethoxysilane.



**Figure 2** FTIR spectra of PP (A) before and (B) after grafting reactions with 5 phr of vinyl trimethoxysilane.



**Figure 3** FTIR spectra of LLDPE (A) before and (B) after grafting reactions with 5 phr of vinyl trimethoxysilane.

### FTIR Analyses

Silane grafting reactions of LDPE, LLDPE, and PP were characterized by FTIR (Bio-Rad FTS 155) in the range of 400–4000  $\text{cm}^{-1}$  with a resolution of 2  $\text{cm}^{-1}$ .

### DSC Measurements

Samples were heated in DSC (TA Instruments, New Castle, DE, DSC 2010) at a rate of 10°C/min from 30 to 150°C (or 200°C for the sample involving PP) holding 3 min, followed by cooling at 2°C/min to 30°C, and then heating again at 2°C/min to 150°C (or 200°C) to obtain the first cooling exotherms and the second heating endotherms.

### Polarized Optical Microscopy

Film samples prepared by casting from xylene were heated in a hot stage (Mettler, FP900) under

nitrogen to 180°C holding for 5 min and were then cooled at a rate of 10°C/min to 100°C holding for 10 h for complete crystallizations. The treated samples were then cooled at 10°C/min to 30°C and transferred to a polarized optical microscope (POM, Zeiss D-07740) to observe their crystal morphologies. The samples characterized by POM included LDPE/LLDPE = 100/0, 75/25, 50/50, 25/75, and 0/100 blends.

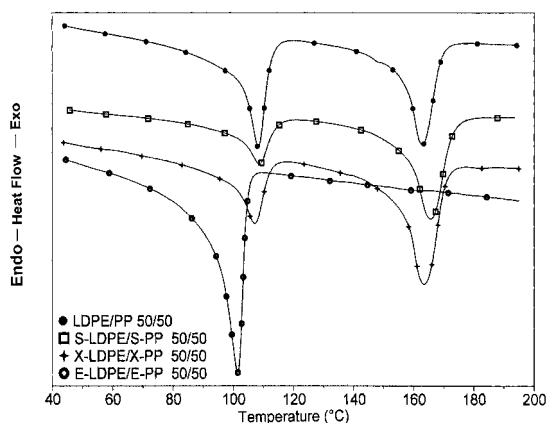
## RESULTS AND DISCUSSION

Figures 1–3 show FTIR spectra of LDPE, PP, and LLDPE, respectively, for both before and after reactions with 5 phr of vinyl trimethoxysilane. As can be seen in Figures 1–3, all three polymers give at least one new peak at 1092  $\text{cm}^{-1}$  after reactions with the silane. The peak at 1092  $\text{cm}^{-1}$  is assigned to the Si—OCH<sub>3</sub> group,<sup>11,13,14</sup> an indication that silane grafting reactions have occurred in these three polymers. Table II lists degrees of crosslinking for S-LDPE/S-PP = 100/0, 50/50, and 0/100 blends after crosslinking reactions in water at various conditions. As can be seen in Table II, S-PP (namely, S-LDPE/S-PP = 0/100) gives negligible degree of crosslinking for reaction conditions studied. Although PP can be silane-grafted as demonstrated in Figure 2, PP can also be easily degraded by heat due to lots of tertiary carbons on the backbone of the polymer. The hydrogen attached to the tertiary carbon is believed<sup>11,12,15</sup> to be easily abstracted to proceed not only the free radical-induced silane grafting reactions but also the thermal degradation

**Table II** Degrees of Crosslinking<sup>a</sup> for S-LDPE/S-PP = 100/0, 50/50, 0/100 Blends after Crosslinking Reactions in Water at Various Conditions

Crosslinking Conditions		S-LDPE/S-PP 100/0	S-LDPE/S-PP 50/50	S-LDPE/S-PP 0/100
100°C	6 h	12.6%	—	—
	12 h	11.0%	—	—
	24 h	34.2%	14.0%	1.6%
120°C	6 h	34.2%	—	—
	12 h	43.3%	—	—
	24 h	—	13.0%	1.1%
140°C	6 h	52.0%	—	—
	12 h	50.0%	—	—
150°C	4 h	37.2%	20.0%	1.5%
170°C	4 h	38.5%	20.6%	1.3%

<sup>a</sup> All values listed in this table are an average value from two experimental data with a deviation less than 10%.



**Figure 4** DSC thermograms of the second heating scans for LDPE/PP = 50/50, S-LDPE/S-PP = 50/50, X-LDPE/X-PP = 50/50, and E-LDPE/E-PP = 50/50 blends.

through the involvement of oxygen dissolved in water at elevated temperatures via a free radical mechanism. In fact, discoloration is observed for S-PP after crosslinking reactions have been proceeded at 150 and 170°C, indicating that degradation of PP really occurs at these temperatures. For S-LDPE (namely, S-LDPE/S-PP = 100/0), considerable degrees of crosslinking are observed at 100°C for 24 h or at reaction temperatures over 100°C for 6 h or longer. For S-LDPE/S-PP = 50/50 blend, the degrees of crosslinking at reaction conditions studied are almost one-half of those for S-LDPE, suggesting that the crosslinking occurs only in the S-LDPE component of the S-LDPE/S-PP = 50/50 blend. Figure 4 shows DSC thermograms of the second heating scans for the LDPE/PP = 50/50 blend for before and after grafting reaction, crosslinking reaction, and decalin extraction. From the curve for E-LDPE/E-PP

= 50/50, the peak at around 163°C is missing, an indication that S-PP gives negligible crosslinking reaction. DSC data in Figure 4 is, thus, consistent with degree of crosslinking data as listed in Table II.

Table III lists degrees of crosslinking for S-LDPE/S-LLDPE = 100/0, 75/25, 50/50, 25/75, and 0/100 blends after crosslinking reactions in water at various conditions. As can be seen in Table III, S-LLDPE gives higher degrees of crosslinking than S-LDPE for all reaction conditions studied. The degrees of crosslinking of S-LDPE/S-LLDPE blends are not linearly proportional to the S-LDPE or the S-LLDPE content in the blends, suggesting that the degrees of crosslinking of S-LDPE/S-LLDPE blends are dependent upon phase separation behavior and morphologies of the constituents of the blends.

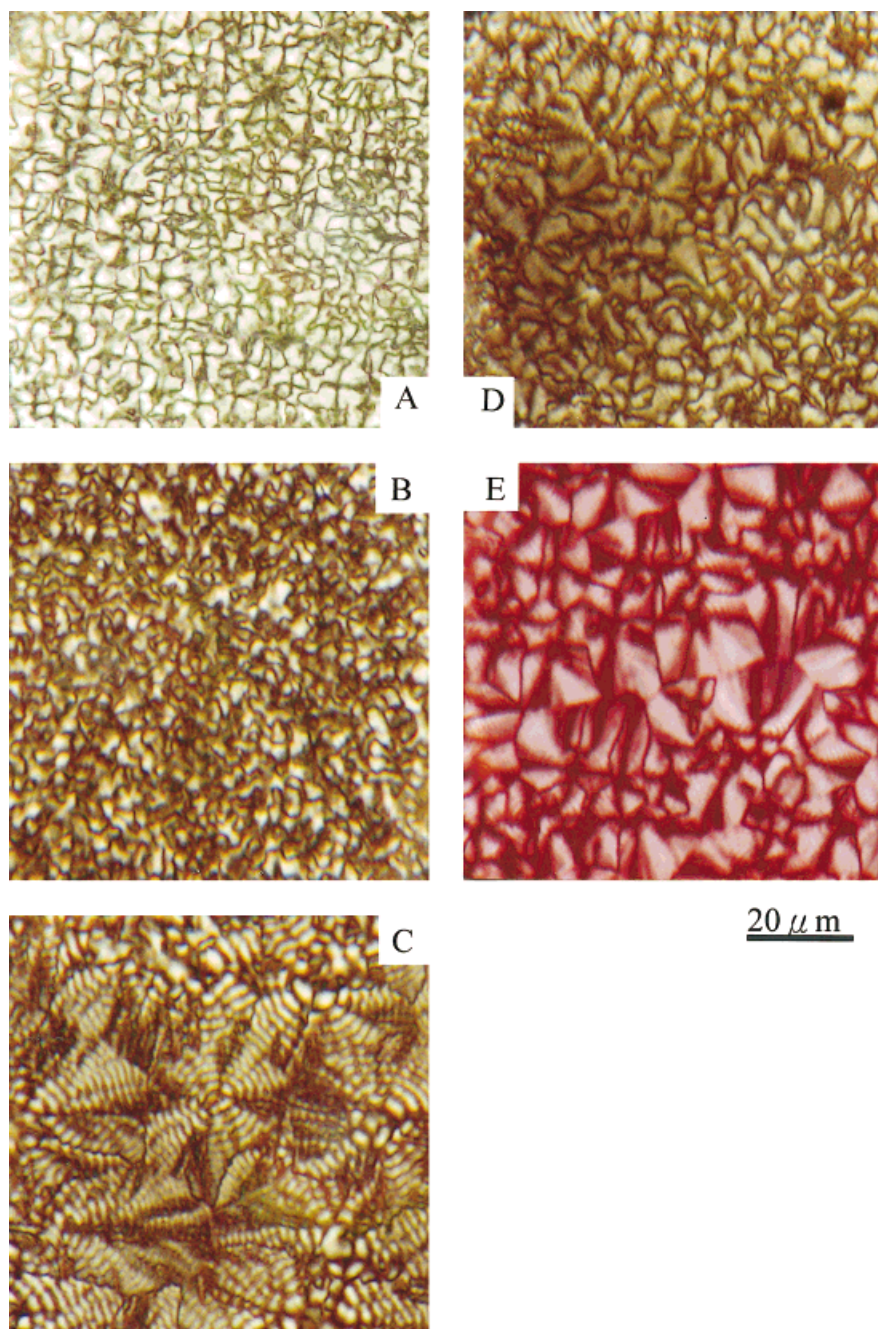
Figure 5 shows polarized optical micrographs for LDPE/LLDPE = 100/0, 75/25, 50/50, 25/75, and 0/100. Spherulite formations can be seen from these micrographs for all five samples. As can be seen from micrograph A in Figure 5 for LDPE, there are many spherulites with a varied size impinging on each other. In micrograph E for LLDPE, the spherulites are much bigger than those for LDPE. In micrograph B for LDPE/LLDPE = 75/25, the spherulites are smaller than those for LDPE because LLDPE crystallizes before LDPE and the crystallized LLDPE acts as a nucleating agent in the crystallization of LDPE, resulting in a decreased spherulite size for LDPE. The spherulites in micrographs C and D are much heterogeneous in size for LDPE/LLDPE = 50/50 and 25/75 blends, respectively, apparently due to phase separation of the two constituents of the blends.

**Table III** Degrees of Crosslinking<sup>a</sup> for S-LDPE/S-LLDPE = 100/0, 75/25, 50/50, 25/75, 0/100 Blends after Crosslinking Reactions in Water at Various Conditions

Crosslinking Conditions	S-LDPE/S-LLDPE					
	100/0	75/25	50/50	25/75	0/100	
100°C	6 h	12.6%	6.9%	24.0%	7.5%	15.4%
	12 h	11.0%	9.5%	17.7%	15.0%	48.0%
	24 h	34.2%	13.5%	34.2%	17.9%	58.5%
120°C	6 h	34.2%	14.0%	43.0%	6.5%	62.4%
	12 h	43.3%	20.0%	40.0%	25.0%	63.0%
140°C	6 h	52.0%	11.9%	40.0%	30.7%	62.5%
	12 h	50.0%	27.4%	27.0%	38.3%	63.5%

<sup>a</sup> All values listed in this table are an average value from two experimental data with a deviation less than 10%.

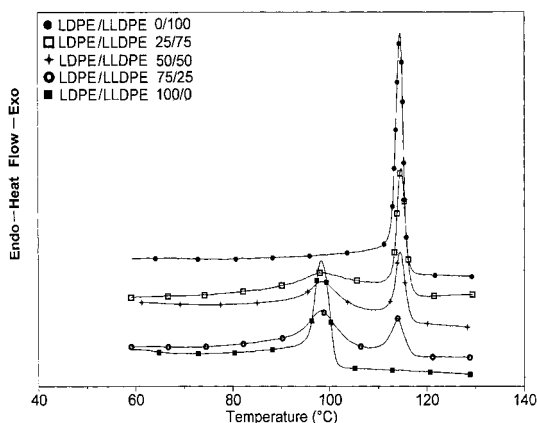




**Figure 5** Polarized optical micrographs observed from crystallization at 100°C for 12 h of the LDPE/LLDPE blends with content ratios at (A) 100/0, (B) 75/25, (C) 50/50, (D) 25/75, and (E) 0/100. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

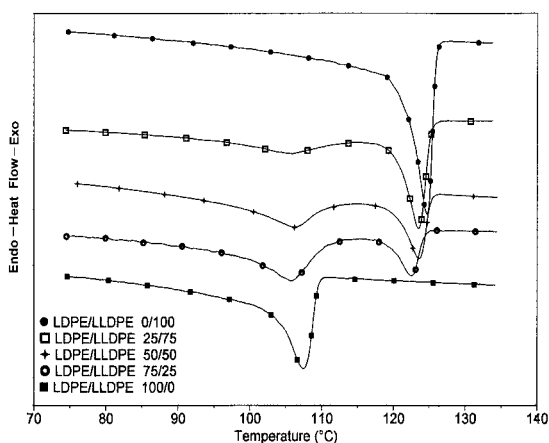
Phase separation of LDPE and LLDPE is also demonstrated by DSC measurements. Figure 6 shows DSC thermograms of the first cooling scans for the LDPE/LLDPE blends. As can be seen in Figure 6, for pure LDPE and LLDPE, crystallization temperatures are around 98 and 114°C, re-

spectively. These crystallization temperatures remain seen for LDPE/LLDPE = 75/25, 50/50, and 25/75 blends, suggesting that cocrystallization of the LDPE and LLDPE in the blends does not occur. In other words, LDPE phase separates from LLDPE phase in the blends. Figure 7 shows

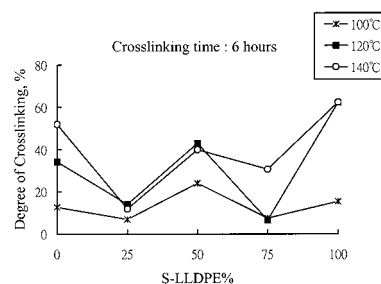


**Figure 6** DSC thermograms of the first cooling scans for the LDPE/LLDPE = 100/0, 75/25, 50/50, 25/75, and 0/100 blends.

DSC thermograms of the second heating scans for the LDPE/LLDPE blends and demonstrates two melting endotherms for the blends with one peak at around 107°C for LDPE and the other one at around 125°C for LLDPE, indicating the presence of two phases in the blends. The peak widths of LDPE and LLDPE exotherms in Figure 6 are bigger for the polymers in the blends than for pure polymers. Although cocrystallization of the LDPE and the LLDPE does not occur, the two polymers in the blends apparently affect crystallizations of each other due perhaps to entanglement and/or interaction between molecules, suggesting that these two polymers are melt miscible and are, thus, compatible in an amorphous domain. The present effect on crystallization of one constituent by the other in the blends is also



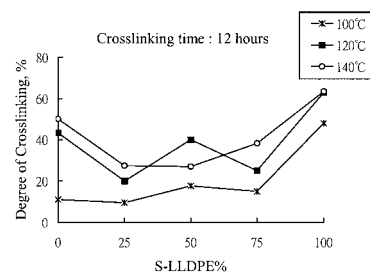
**Figure 7** DSC thermograms of the second heating scans for the LDPE/LLDPE = 100/0, 75/25, 50/50, 25/75, and 0/100 blends.



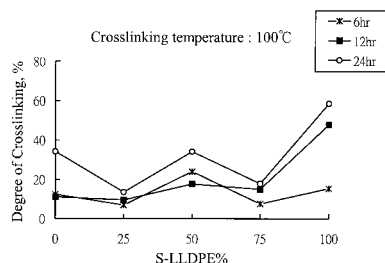
**Figure 8** Degrees of crosslinking of the S-LDPE/S-LLDPE blends as functions of the S-LLDPE contents after crosslinking for 6 h at 100, 120, and 140°C.

demonstrated by the second heating endotherms in Figure 7 where the melting temperature of one constituent is found to decrease with increasing content of the other constituent.

Figures 8–10 are plotted from data listed in Table III. As can be seen for any crosslinking condition, the degree of crosslinking of S-LLDPE is higher than that of S-LDPE. The degrees of crosslinking are not linearly but are zigzagly related to the compositions of the blends, suggesting that both silane-grafted polymers exhibit different crosslinking rates in comparison between these polymer in the blends and in the pure state. These changes in crosslinking rates of the polymers after blending apparently result from phase separation of these two polymers in the blends. Compared with S-LDPE/S-LLDPE = 75/25 and 25/75 blends, the S-LDPE/S-LLDPE = 50/50 blend gives a relatively high degree of crosslinking for almost all crosslinking conditions studied except the one at 140°C for 12 h as shown in Figure 9. This can be explained in terms of the phase behavior of the blends and its dependence on temperature and composition. For the blends at 100 and 120°C, LLDPE exhibits a slower chain mobility than LDPE because melting temperature of LLDPE is 125°C, and is higher than that of



**Figure 9** Degrees of crosslinking of the S-LDPE/S-LLDPE blends as functions of the S-LLDPE contents after crosslinking for 12 h at 100, 120, and 140°C.



**Figure 10** Degrees of crosslinking of the S-LDPE/S-LLDPE blends as functions of the S-LLDPE contents after crosslinking at 100°C for 6, 12, and 24 h.

LDPE at 107°C (Fig. 7). For the LDPE-rich blend (namely, LDPE/LLDPE = 75/25) at 100 and 120°C, LDPE is a continuous phase, whereas LLDPE is a dispersed phase (or discontinuous phase). For the LLDPE-rich blend (namely, LDPE/LLDPE = 25/75) at 100 and 120°C, LLDPE is a continuous phase, whereas LDPE is a dispersed phase. For the LDPE/LLDPE = 50/50 blend at 100 and 120°C, LDPE and LLDPE are cocontinuous phases. These phase behavior is also previously demonstrated in polarized optical micrographs as shown in Figure 5. Thus, at 100 and 120°C, the less crosslinking ability of S-LDPE and the slower chain mobility of S-LLDPE result in the finding of a relatively high degree of crosslinking for the S-LDPE/S-LLDPE = 50/50 blend compared with that for the S-LDPE/S-LLDPE = 75/25 or 25/75 blend.

At 140°C, the two constituents in the blends are in the melting state. The molecular weight, or the viscosity, of the polymer should be taken into consideration, a main factor to determine its ultimate degree of crosslinking. The higher in the chain mobility, namely, the lower in molecular weight, gives the higher in the crosslinking rate and thus the degree of crosslinking. As can be seen in Figure 9, for 12 h of crosslinking at 140°C, the S-LDPE/S-LLDPE = 50/50 blend gives a less degree of crosslinking than S-LDPE/S-LLDPE = 75/25 and 25/75 do, although a different result is obtained for a shorter time (6 h) of reaction, as shown in Figure 8. For 12 h at 140°C at which all three blends are in melting state, the reason for the S-LDPE/S-LLDPE = 50/50 blend giving the lowest degree of crosslinking among the blends studied is perhaps as follows: In LDPE-rich blend, the LDPE (MI 2.8) with the lower molecular weight, or the higher chain mobility, dominates the crosslinking reaction, although the LDPE has the less crosslinking ability. In LLDPE-rich blend, the LLDPE with the higher crosslinking

ability dominates the crosslinking reaction, although the LLDPE (MI 0.8) has the lower chain mobility.

For 100°C at which the LLDPE in the blends is in its crystalline state, as can be seen in Figure 10, degrees of crosslinking of the S-LDPE/S-LLDPE = 75/25 and 25/75 blends are insignificantly functions of crosslinking times. In the LDPE-rich blend at 100°C, the LLDPE is a dispersed phase and, thus, gives insignificant contribution to the increase of degree of crosslinking while the LDPE with the less crosslinking ability reaches its ultimate extent of crosslinking reaction in 6 h and, thus, exhibits only a slight increase with increasing times. In the LLDPE-rich blend at 100°C, the LLDPE is in its crystalline state and is a continuous phase with LDPE dispersing in the LLDPE phase and, thus, gives a significantly reduced ability to the increase of degree of crosslinking leading to a weak dependence on crosslinking times for the degree of crosslinking of the blend.

## CONCLUSIONS

This article aims at investigation of crosslinking reactions of LDPE, PP, LLDPE, and the binary blends of these three polyolefins. Quantitative comparisons are conducted and discussed on degrees of crosslinking of these samples in terms of crosslinking ability, phase separation behavior, molecular weight (or viscosity), morphology of the constituents in blends, or in the pure state. PP is found to give a negligible degree of crosslinking in the pure state and in blends with LDPE. LDPE and LLDPE are both capable of giving a considerable degree of crosslinking in the pure state and in blends of both polymers. Degrees of crosslinking of the LDPE/LLDPE blends are dependent upon temperature and time of crosslinking reactions and morphology and composition of the blends.

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