Functionalization of Polyolefins and Elastomers with an Oxazoline Compound

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ABSTRACT: The aim of this work was to graft ricinoloxazoline maleinate (OXA) onto polyethylene (PE), and onto an ethylene propylene copolymer (E/P) and styrene ethylene/butylene styrene copolymer (SEBS), by melt free radical grafting in a twin-screw midiextruder. A study was made of the effects of the initial monomer and peroxide concentrations and of temperature on the degree of grafting, on the amount of residual monomer, and on the molecular weight. The initial monomer and peroxide concentrations were 1.5–9.0 wt % and 0.15–0.90 wt %, respectively. The grafting yield was found to increase with the initial monomer and peroxide concentrations. Grafting yields up to 2.1 wt % for PE, 2.3 wt % for E/P and 2.7 wt % for SEBS were achieved. The degree of grafting also varied considerably with the temperature. E/P and SEBS decomposed with low initial monomer and peroxide concentrations, but crosslinked with raising the initial concentrations. Polyethylene crosslinked even with low initial concentrations, but with a suitable choice of peroxide grafted polyethylene could be produced with good grafting yields and without gel formation. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 72: 877–885, 1999

Key words: oxazoline; melt free radical grafting; reactive extrusion; polyolefin; styrene ethylene/butylene styrene copolymer

INTRODUCTION

The development of new polymer blends is an attractive area of research because it is more economical to produce new materials by blending existing polymers than by synthesizing new ones. Most thermoplastic polymers are, however, immiscible and often poorly compatible with each other, which means that a third component is needed to increase the compatibility of the blend components. The most common compatibilizers are block and graft copolymers, which are presumed to locate at the blend interface, reducing the interfacial tension, increasing the adhesion between the separate phases, promoting the dispersivity, and controlling the particle size of the dispersed phase.[1,2]

Graft copolymers of polyolefins are widely used as compatibilizers in blends of polyolefins with engineering plastics such as polyamides and polyesters. Compared with neat polyolefins, which are nonpolar, the functionalized polyolefins exhibit better adhesion, dyeability, paintability, and printability. Functionalized copolymer compatibilizers can be produced by melt free radical grafting through reactive extrusion. Compared with grafting in solution, melt free radical grafting is more advantageous in terms of energy and the environment because no solvent is needed, and therefore, no solution waste is produced.[3] The grafted product is also ready for use right after the extrusion process, or grafting and blending

can even be performed *in situ*, all in one extrusion run.[3,4]

The most frequently used monomer for grafting polyolefins and elastomers is maleic anhydride (MA). Polymers functionalized with MA are widely used as effective compatibilizers in polymer blends. Other common functional monomers are glycidyl methacrylate (GMA) and carboxylic acids.[5] Likewise, oxazoline compounds are interesting monomers for producing compatibilizers by melt free radical grafting. Long-chain oxazolines have been reported to be less toxic than maleic anhydride and glycidyl methacrylate, and their boiling points are well above those of MA and GMA.[6,7] Oxazolines react fast with carboxyl and amino groups, which makes them suitable for use in blends containing these end groups.[8–10] They also react with other functional groups such as phenol and mercaptan.[11]

Polyethylene and polypropylene have previously been grafted with various oxazoline compounds. Vainio et al.[9] grafted ricinoloxazoline maleinate onto polypropylene by reactive extrusion, achieving a grafting degree of 0.4 to 2.1 wt %. The grafted product was found to work well as a compatibilizer for PP/PBT blends. Liu et al.[12,13] prepared a functional polypropylene by melt free radical grafting of isopropenyloxazoline onto the polymer, and used the product as a compatibilizer for polypropylene/(acrylonitrile/butadiene/acrylic acid) blends. The compatibilizer decreased the size of the dispersed particles and improved the impact strength of the blend. The grafting of polyethylene with different oxazoline monomers in a twin screw extruder has been screened by Birnbrich et al.[14,15]

In this work we studied the melt free radical grafting of ricinoloxazoline maleinate (OXA) onto polyethylene, ethylene propylene copolymer (E/P) and styrene ethylene/butylene styrene (SEBS) copolymer. The structure of OXA is depicted in Figure 1. The grafting reaction was examined by FTIR and ^{1}HNMR analyses.

EXPERIMENTAL

Materials

The polyethylene was LE7518 (density 920 kg/m^3), supplied by Borealis Polymers; the ethylene propylene copolymer was Hifax CA10A, supplied by Himont; and the styrene ethylene/butylene

Figure 1   Ricinoloxazoline maleinate.

styrene block copolymer was Kraton G-1652 (styrene content 30 wt %), supplied by Shell. The functional monomer was ricinoloxazoline maleinate available as Loxamid V-EP 8515 (Henkel KGaA). This monomer was in liquid form and exhibited very low volatility (bp > 250°C at 0.1 mbar). The peroxide initiators were 1,1-bis(*tert*-butylperoxy)-3,3,5-trimethylcyclohexane, Trigonox 29-C90 (T29), for the grafting onto polyethylene; cumyl hydroperoxide, Trigonox K-95 (TK), for SEBS; and bis(*tert*-butylperoxyisopropyl)benzene, Perkadox 14S-fl (P14), for polyethylene, E/P copolymer and SEBS, all supplied by Akzo.

Grafting Procedure

The melt free radical grafting was carried out in a corotating twin-screw midiextruder (DSM, capacity = 16 cm^3, screw length L = 150 mm). The midiextruder can be used for both batch and continuous mixing. The screw speed was 50 rpm during filling and 65 rpm during mixing. Before the material was fed into the extruder, the monomer and peroxide initiator were allowed to absorb into the matrix material, which was in powder form. The absorption procedure was carried out at room temperature for at least 15 min. The grafting time was 5 min, after which the material was taken out of the extruder and immediately cooled in a cold water bath. The grafting was made under nitrogen atmosphere. The initial concentration of oxazoline monomer (OXA$_i$) varied between 1.5 and 9 wt %, and that of the peroxide (ROOR$_i$) between 0.15 and 0.9 wt %. The grafting temperature was chosen between 170 and 260°C, depending on the matrix material.

Purification of Samples and Characterization

The functionalized samples were purified by dissolving them in boiling decane (polyethylene) or xylene (E/P and SEBS) and precipitating them

with acetone at room temperature. With use of this procedure the free oxazoline and its possible homopolymers were separated from the grafted polymer, and the grafting yield was determined. Some polyethylene samples did not dissolve completely, which indicated gel formation. Undissolved samples were not further characterized, because polymers containing gel fractions would not be as effective in compatibilization or adhesion studies as polymers without gel.

The relative grafting yield was determined by FTIR (Nicolet Magna 750) from thin compression moulded films of the purified samples. The peak at 1671 cm^{-1}, attributed to a complex mode involving C=N stretching and oxazoline ring bending, was treated as a characteristic peak of the grafted ricinoloxazoline maleinate. Another peak at 1737 cm^{-1}, resulting from the ester group in the oxazoline monomer, was used to verify the results. For polyethylene the peak at 1464 cm^{-1} representing the methylene groups was chosen as reference, and the peak at 720 cm^{-1}, also due to the methylene groups, was used to verify the results. For the E/P copolymer the peak at 2723 cm^{-1} representing the skeleton of the PP chain was chosen as reference, and to verify the results the peak at 1460 cm^{-1} was studied. For the SEBS copolymer the peak at 1601 cm^{-1} was chosen as reference, and the peak at 1493 cm^{-1} to verify the results. Both peaks were due to the styrene blocks of the copolymer.

^{1}HNMR analysis (Varian Gemini 2000, 300 MHz, BB) was used to calculate precise grafting yields, and to produce a calibration curve for the IR results. The samples were dissolved in deuterated tetrachloroethane at 125°C. The protons of the oxazoline and ester groups of OXA produced peaks in the 3.5–3.9 and 4.1–4.3 ppm regions. Many other peaks, resulting from the complex structure of the monomer, were also observed, but omitted in the calculations. The protons of polyethylene, the E/P copolymer, and the butylene blocks of SEBS copolymer produced peaks in the 0.8–1.8 ppm region, while the styrene blocks of SEBS produced peaks in the 7.3–6.3 ppm region. The grafting yields were calculated from the peaks of the hydrogens in the oxazoline ring and the peaks in the polyolefin region.

The melt flow index (MFI) was measured according to standard ISO 1133-1981(E) with a Ceast melt flow index measurement device under a load of 5000 g at 190°C for polyethylene, 200°C for E/P, and 250°C for SEBS.

RESULTS AND DISCUSSION

Effect of Initial Monomer Concentration on the Grafting Yield

The grafting yields (OXA$_g$), amounts of residual monomer (OXA$_r$), grafting efficiencies (GE), and melt flow indices (MFI) for E/P and SEBS are set out in Table I, and the same values for polyethylene are given in Table II. The grafting efficiency is defined as the amount of grafted monomer relative to the total amount of monomer in the extrudate. If the polyethylene sample did not dissolve in boiling decane and was thereby assumed to include gel fractions, the monomer contents and melt flow indices were not determined.

The effect of the initial monomer concentration on the grafting yield and on the residual amount of OXA is illustrated in Figure 2. The peroxide concentration was kept at 10 wt % of the monomer concentration for E/P and SEBS and at 6.7 wt % for PE. The grafting temperature was 200°C for E/P, 250°C for SEBS, and 170°C for PE. For E/P and SEBS the peroxide used was P14, and for polyethylene T29.

As expected, the higher the initial monomer concentration the higher was the grafting yield. For example, when the initial monomer concentration was increased from 3 to 9 wt %, the grafting yield increased from 0.7 to 2.3 wt % for E/P, to 2.7 wt % for SEBS and from 0.6 to 2.1 wt % for PE. Simultaneously, the amount of residual OXA increased. For E/P and SEBS the grafting yield was substantially higher than the residual amount of OXA at initial monomer concentrations above 4.5 wt %. At lower initial concentrations the grafted and residual amounts were more or less equal. In the case of polyethylene the grafting yield exceeded the residual monomer amount at all initial monomer concentrations.

Effect of Temperature and Initiator Type on the Grafting Yield

Initiators were selected so as to have a half-life of less than 1 min in the temperature range tested. For E/P the temperature was varied between 180 and 200°C, for SEBS between 240 and 260°C, and for PE between 170 and 190°C. The half-lives of the peroxides at different temperatures were extrapolated from the half-life and the Arrhenius equations:[16]

Table I   Grafting Yield (OXA$_g$), Amount of Residual Monomer (OXA$_r$), Grafting Efficiency (GE), and Melt Flow Index (MFI) of E/P and SEBS

Material	T (°C)	OXA$_i$ (wt %)	ROOR$_i$ (wt %)	OXA$_g$ (wt %)	OXA$_r$ (wt %)	GE (%)	MFI (g/10 min)
E/P	200	—	—	—	—	—	1.5
E/P	200	1.5	0.15	0.4	0.3	62	3.7
E/P	200	3.0	0.30	0.7	0.6	56	3.5
E/P	200	3.0	0.60	0.9	0.4	67	6.7
E/P	200	4.5	0.45	0.9	0.9	50	1.4
E/P	200	6.0	0.30	1.1	1.3	45	0.3
E/P	200	6.0	0.60	1.4	1.0	58	0.6
E/P	200	6.0	0.90	1.7	0.7	70	0.4
E/P	200	9.0	0.60	1.9	1.8	51	0.1
E/P	200	9.0	0.90	2.3	1.4	62	<0.1
E/P	190	6.0	0.60	1.3			
E/P	170	6.0	0.60	1.2			
SEBS	250	—	—	—	—	—	14.2
SEBS, P14	250	1.5	0.15	0.3	0.3	48	26.6
SEBS, P14	250	3.0	0.30	0.7	0.6	53	10.9
SEBS, P14	250	3.0	0.60	0.8	0.4	64	1.0
SEBS, P14	250	4.5	0.45	1.1	0.9	56	4.0
SEBS, P14	250	6.0	0.30	1.2	1.6	43	7.5
SEBS, P14	250	6.0	0.60	1.4	0.9	61	3.5
SEBS, P14	250	6.0	0.90	1.8	0.9	68	0.1
SEBS, P14	250	9.0	0.60	2.2	2.1	52	0.4
SEBS, P14	250	9.0	0.90	2.7	1.5	65	<0.1
SEBS, P14	260	6.0	0.60	1.5			
SEBS, P14	240	6.0	0.60	1.9			
SEBS, TK	250	3.0	0.3	0.3			
SEBS, TK	250	6.0	0.6	0.6			

$$t_{1/2} = \ln 2/k_d \qquad (1)$$

$$k_d = A \cdot e^{-E_a/RT} \qquad (2)$$

where $t_{1/2}$ is the half-life, k_d the rate constant for the initiator dissociation, A the Arrhenius frequency factor, E_a the activation energy for the initiator dissociation, R the gas constant, and T the absolute temperature.

The Arrhenius frequency factors, the activation energies and the calculated half-lives for the peroxides are presented in Table III. In setting up the experiments it was supposed that P14 was a suitable initiator for the grafting onto E/P because of its appropriate half-life. It was also supposed that TK would be more suitable for the grafting onto SEBS, owing to the fast decomposition of P14 at 240–260°C. For polyethylene, on the other hand, it was expected that P14 would decompose too slowly at 170°C, causing undesired residual peroxide in the extrudate.

The effects of temperature and initiator type on grafting yields of OXA for E/P and SEBS are displayed in Figure 3. The initial monomer concentration was 6 wt %, and that of peroxide 0.6 wt %. As can be seen, for E/P, a rise in temperature, and consequently a decrease in half-life of the peroxide, caused an increase in the grafting yield. Higher temperatures than 200°C were not tested because the process should be performed close to the melt temperature of the matrix material to avoid excessive degradation. For SEBS, P14 turned out to be a more suitable initiator than TK, despite its very fast degradation. The grafting yields achieved with TK were only half of those achieved with P14 (Table I). TK is a hydroperoxide, which seems to limit the grafting reaction to some extent. The highest grafting yield with P14 was achieved at 240°C. However, the processing of SEBS is more difficult at such a low temperature and, therefore, a temperature of 250°C was chosen for the grafting.

Table II   Grafting Yield (OXA$_g$), Amount of Residual Monomer (OXA$_r$), Grafting Efficiency (GE), Melt Flow Index (MFI) of Polyethylene

ROOR	T (°C)	OXA$_i$ (wt %)	ROOR$_i$ (wt %)	OXA$_g$ (wt %)	OXA$_r$ (wt %)	GE (%)	MFI (g/10 min)
—	170	—	—	—	—	—	10.3
T29	170	1.5	0.15	0.3	0.2	58	1.9
T29	170	3.0	0.15	0.5	0.6	46	0.5
T29	170	3.0	0.20	0.6	0.5	57	0.2
T29	170	3.0	0.30	0.6	0.5	56	0.1
T29	170	4.5	0.15	0.7	1.0	40	0.3
T29	170	4.5	0.225	0.9	0.8	50	0.1
T29	170	4.5	0.30	0.9	0.7	55	<0.1
T29	170	4.5	0.45	1.0	0.6	62	<0.1
T29	170	4.5	0.60	cross	linked		
T29	170	6.0	0.15	0.9	1.2	42	0.4
T29	170	6.0	0.30	1.0	1.2	46	0.1
T29	170	6.0	0.45	1.2	1.0	54	<0.1
T29	170	6.0	0.60	cross	linked		
T29	170	9.0	0.15	1.1	2.4	31	0.6
T29	170	9.0	0.30	1.3	1.9	42	0.1
T29	170	9.0	0.60	2.1	1.4	60	<0.1
T29	170	9.0	0.90	cross	linked		
P14	170	1.5	0.15	0.2	0.3	45	1.4
P14	170	3.0	0.15	0.5	0.5	49	0.4
P14	170	3.0	0.20	0.6	0.6	49	0.4
P14	170	3.0	0.30	0.6	0.5	55	<0.1
P14	170	4.5	0.15	0.7	1.0	42	0.3
P14	170	4.5	0.225	cross	linked		
P14	170	6.0	0.15	0.9	1.4	39	0.6
P14	170	6.0	0.30	cross	linked		
P14	170	9.0	0.15	1.2	2.4	33	0.2
P14	170	9.0	0.30	cross	linked		
P14	190	1.5	0.15	0.3	0.2	61	1.3
P14	190	3.0	0.15	0.6	0.6	49	0.2
P14	190	3.0	0.20	cross	linked		
P14	190	4.5	0.15	0.9	1.1	46	0.2
P14	190	4.5	0.225	cross	linked		
P14	190	6.0	0.15	1.1	1.2	50	0.2
P14	190	6.0	0.30	cross	linked		
P14	190	9.0	0.15	1.5	2.1	42	<0.1
P14	190	9.0	0.30	cross	linked		

The grafting yields of OXA on polyethylene at different temperatures and with both peroxides are displayed in Figure 4. The peroxide content was kept constant at 0.15 wt %. A well-known problem in grafting onto polyethylene is crosslinking. Increasing the initial peroxide concentration only slightly above 0.15 wt % at 190°C with P14 resulted in undissolved fractions. Reduction of the temperature to 170°C allowed grafting with somewhat higher initial peroxide concentrations, but grafting yields were clearly lower. With the peroxide T29, with a shorter half-life, about 10 s at 170°C, undissolved fractions appeared only when substantially higher initial concentrations were applied. The peroxides used in this work resulted in fairly similar grafting yields. An increase in temperature, on the other hand, led to increased grafting yields, but unfortunately the sensitivity to crosslinking increased as well. The peroxide T29 did not induce crosslinking as readily as P14, and it could therefore be used at higher initial peroxide concentrations.

Figure 2 Effect of the initial monomer concentration on the grafting yield (filled) and residual amount (open) of OXA for polyethylene ($\square$,$\blacksquare$), E/P ($\blacktriangle$,$\triangle$), and SEBS ($\bullet$,$\circ$). T = 170°C for PE, 200°C for E/P and 250°C for SEBS. $ROOR_i$/OXA_i = 6.7 wt % for PE and 10 wt % for E/P and SEBS.

Effect of Initial Peroxide Concentration on the Grafting Yield

The effect of the initial peroxide concentration on the grafting yield, and on the residual amount of OXA for E/P and SEBS, with an initial monomer concentration of 6.0 wt % is illustrated in Figure 5. As expected, the grafting yield increased with the initial peroxide concentration. Simultaneously, the residual amount of OXA decreased, and both the conversion and grafting efficiency increased. The corresponding values for polyethylene with initial monomer concentrations of 4.5 and 9.0 wt % are depicted in Figure 6. Again, as the initial peroxide concentration was increased, the grafting yield increased and the amount of residual monomer decreased. Consequently, the conversion and the grafting efficiency increased also. Conversions of about 30% were achieved for E/P and SEBS. Because of easy crosslinking of polyethylene, low initial peroxide to monomer ratios had to be used, resulting in conversions of 23% at maximum.

Melt Flow Index of Functionalized Polymers

Results of melt flow index (MFI) measurements are depicted in Table I for E/P and SEBS, and in Table II for polyethylene. Well-known difficulties are crosslinking in grafting onto polyethylene and β-scission in grafting onto polypropylene. For polyethylene, a significant decrease in MFI was observed even with low monomer and peroxide additions, which indicates high extent of crosslinking. Both peroxides strongly induced crosslinking, but with the peroxide T29 higher initial peroxide concentrations could be used to still get processable products, that is, products with crosslinking, but without gel.

Table III Arrhenius Frequency Factors (A), Activation Energies (E_a) and Half-Lives ($t_{1/2}$) for the Peroxides Used

Peroxide	T (°C)	A (1/s)	E_a (J/mol)	$t_{1/2}$ (s)
P14		$7.65 * 10^{15}$	152.69	
	170			90
	180			36
	190			15
	200			7
	240			0.3
	250			0.2
	260			0.1
TK		$1.15 * 10^{12}$	132.56	
	250			10
T29		$7.59 * 10^{13}$	127.52	
	170			10

Figure 3 Effect of temperature on the grafting yield of OXA for E/P (▲) and SEBS (●, ◐) with P14 (black) and TK (gray), $OXA_i = 6$ wt % and $ROOR_i = 0.6$ wt %.

In grafting onto E/P or SEBS the peroxide radical is assumed to abstract a tertiary hydrogen from a polymer backbone. Thus, if grafting does not occur, the most predictable reaction is β-scission. With low initial peroxide and monomer concentrations in grafting onto E/P or SEBS, the MFI increased as a result of β-scission. In raising the initial concentrations the MFI gradually decreased as a result of crosslinking. This behavior is assumed to result from polymer radicals formed from abstraction of secondary hydrogens. In grafting onto SEBS, crosslinking overcame β-scission with lower monomer and peroxide concentrations than in grafting onto E/P, likely be-

Figure 4 Effect of the initial monomer concentration on the grafting yield (filled) and residual amount (open) of OXA for PE at different temperatures and with both peroxides, P14 170°C (▲,△), P14 190°C (●,○), and T29 170°C (■,□). $ROOR_i$ was constant at 0.15 wt %.

Figure 5   Effect of the initial peroxide concentration on the grafting yield (filled) and residual amount (open) of OXA for E/P (▲,△) and SEBS (●,○), $T = 200°C$ for E/P and 250°C for SEBS, and $OXA_i = 6.0$ wt %.

cause the amount of tertiary hydrogens is lower in SEBS.

CONCLUSIONS

Ricinoloxazoline maleinate was grafted onto polyethylene, ethylene propylene copolymer, and styrene ethylene/butylene styrene copolymer. Study was made of the effects of the initial monomer and peroxide concentrations and of temperature on the grafting yields and on the residual monomer

amounts. The degree of grafting was found to increase with the initial monomer concentration. At the same time, the amount of residual monomer in the sample increased. The maximum conversions of the ricinoloxazoline maleinate lay at about 30% for E/P and SEBS and at about 23% for polyethylene. Increasing the temperature improved the grafting yield for E/P. An increase in the initial peroxide concentration at a fixed initial monomer concentration increased the grafting yield and simultaneously decreased the residual amount of OXA in the samples. The melt flow index of E/P and SEBS

Figure 6   Effect of the initial peroxide concentration on the degree of grafting (filled) and the amount of residual monomer (open) for polyethylene at 170°C, $[OXA]_i = 4.5$ wt % (■,□) and 9.0 wt % (▲,△).

increased in grafting with low initial monomer and peroxide concentrations, but decreased with higher concentrations, as an indication of crosslinking. Polyethylene crosslinked even with a low initial monomer and peroxide concentrations, but with a suitable choice of peroxide grafted polyethylene could be produced with good grafting yield and without gel formation.

LIST OF SYMBOLS

E/P	ethylene propylene copolymer
E/P-*g*-OXA	oxazoline grafted ethylene propylene copolymer
FTIR	Fourier transferred infrared spectroscopy
GE	grafting efficiency
GMA	glycidyl methacrylate
MA	maleic anhydride
MFI	melt flow index
NMR	nuclear magnetic resonance spectroscopy
OXA	ricinoloxazoline maleinate
PE	polyethylene
PE-*g*-OXA	oxazoline grafted polyethylene
PP	polypropylene
PP-*g*-OXA	oxazoline grafted polypropylene
P14	bis(*tert*-butylperoxyisopropyl)benzene (Perkadox 14S-fl)
ROOR	peroxide
SEBS	styrene ethylene/butylene styrene copolymer
SEBS-*g*-OXA	oxazoline grafted styrene ethylene/butylene styrene copolymer
T	temperature (°C)
TK	cumyl hydroperoxide (Trigonox K-95)
T29	1,1-bis(*tert*-butylperoxy)-3,3,5-trimethylcyclohexane (Trigonox 29-C90)

Subscripts

g	grafted
i	initial, at feed
r	residual, ungrafted

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