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COMPATIBILIZING AGENTS: STRUCTURE AND FUNCTION IN POLYBLENDS[†]

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ABSTRACT

The presence of a compatibilizing agent permits the blending of otherwise incompatible polymers to yield polyblends or alloys with unique properties, generally not attainable from the individual components. Block and graft copolymers possessing segments with chemical structures or solubility parameters which are similar to those of the polymers being blended are effective compatibilizing agents. The latter may be preformed and added to the mixture of polymers undergoing compatibilization or may be generated *in situ* by reaction between coreactive functional groups on the polymers. The bonding between the segments in the compatibilizing agent, whether preformed or generated *in situ*, is generally covalent, ionic, or hydrogen bonding. The compatibilizing agent acts as a polymeric surfactant, lowering surface tension and promoting interfacial adhesion between the dispersed and matrix polymer phases in a polyblend or alloy or between the layers in a laminate formed by extrusion or coextrusion. Numerous polyblends and laminates are discussed as illustrative of the concepts.

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INTRODUCTION

For many years the general incompatibility of polymers was responsible for the inability to prepare useful polyblends. However, in recent years, most of the "new" polymers introduced commercially have actually been blends of older, well-established polymers. The new polyblends or polymer alloys result from the introduction or *in-situ* formation of a compatibilizing agent which permits the blending of otherwise incompatible polymers to yield compositions with unique properties, generally not attainable from either of the components of the polyblend.

DEFINITION OF COMPATIBILITY

In contrast to the concept of thermodynamic compatibility, according to which the chains of two polymers exist homogeneously and intimately mixed on a molecular level, Gaylord [1] defined the term "compatibilizing" in a U.S. patent filed in 1966 as

> "rendering a mixture of two or more polymeric materials permanently miscible so as to form a homogeneous composition which has useful plastic properties and which does not separate into its component parts."

The term "compatibilization" was more clearly defined later by Gaylord [2]:

> "The term 'compatibilization' refers to the absence of separation or stratification of the components of the polymeric alloy during the expected useful lifetime of the product."

A most satisfactory definition was recently provided by Coran and Patel [3] who coined the term "technological compatibilization" and stated

> "Technological compatibilization is the result of a process or technique for improving ultimate properties by making polymers in a blend *less incompatible;* it is *not* the application of a technique which induces 'thermodynamic compatibility' which would cause the polymers to exist in a single molecularly blended homogeneous phase."

FACTORS PROMOTING COMPATIBILITY

Two polymers form a compatible mixture when they possess one or more of the following characteristics:

1. Segmental structural identity
2. Solubility parameter difference less than 1.0
3. Functional groups capable of interaction

Compatible compositions are generated on admixture of polymers which have segmental structural identity, e.g., a graft or block copolymer of butadiene and styrene is compatible with either polybutadiene or polystyrene.

Although they may have different chemical structures, polymers having similar solubility parameters (δ) are generally compatible. Simple organic compounds are compatible, i.e., they form nonseparating mixtures or solutions, when the difference in solubility parameters is as large as 5 units. However, polymer-polymer compatibility requires a difference in solubility parameters of less than 1 unit, generally less than 0.2 units.

Poly(vinyl chloride) (PVC), poly(ethyl acrylate) (PEA), and poly(methyl methacrylate) (PMMA) have solubility parameters in the 9.4-9.5 range and form compatible mixtures. A 70/30 poly(butadiene-*co*-acrylonitrile) has a structure entirely different from PVC, PEA, or PMMA but has a similar solubility parameter of 9.5 and is, therefore, compatible with them.

The presence of functional groups on polymers may promote compatibility as a result of interaction which generates bonding between the polymers. Effective bonds include covalent, ionic, and hydrogen bonds, as well as donor-acceptor interactions.

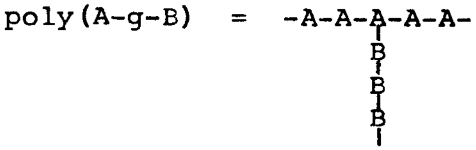
COMPATIBILIZING AGENTS

The seed of the concept of compatibilizing agents may be traced back to the early work on block and graft copolymers. During the preparation of review articles on these polymers whose chains contain sequences of units derived from different monomers, Gaylord [4, 5] noted that most references reported that

> "the block and graft copolymers are not readily characterized since it is difficult to separate them from the mixture of homopolymers."

The products generated from the polymerization of Monomer B in the presence of preformed Polymer A are a mixture of unreacted poly-A, newly formed poly-B from the homopolymerization of monomer B, and a graft copolymer in which a poly-A backbone contains pendent B sequences, referred to as poly(A-*g*-B).

$$\text{Poly-A} \quad + \quad \text{Monomer B} \longrightarrow \begin{pmatrix} \text{poly-A} \\ \text{poly(A-g-B)} \\ \text{poly-B} \end{pmatrix} \tag{1}$$



$$\text{poly(A-g-B)} = -\text{A}-\text{A}-\underset{\substack{| \\ \text{B} \\ | \\ \text{B} \\ | \\ \text{B} \\ |}}{\text{A}}-\text{A}-\text{A}- \tag{2}$$

Analogously, the products generated during the mixing of two homopolymers under shear are a mixture of the two homopolymers and a block copolymer containing sequences of both homopolymers, referred to as poly(A-*b*-B).

$$-\text{A}-\text{A}-\text{A}-\text{A}- \quad + \quad -\text{B}-\text{B}-\text{B}-\text{B}- \longrightarrow \begin{pmatrix} \text{poly-A} \\ \text{poly(A-b-B)} \\ \text{poly-B} \end{pmatrix} \tag{3}$$

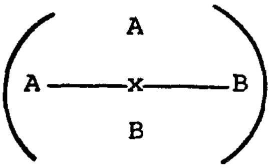
$$\text{poly(A-b-B)} = -\text{A}-\text{A}-\text{A}-\text{B}-\text{B}-\text{B}- \tag{4}$$

It would appear that the presence of the graft or block copolymer inhibits the tendency of the two incompatible homopolymers to separate, i.e., the copolymer promotes compatibility.

In disclosing the compatibilization of a mixture of a nonthermoplastic, though uncrosslinked, polymer such as cellulose with a thermoplastic polymer, Gaylord [1] commented that

> "a cellulose graft copolymer containing segments of a thermoplastic polymer may be used as a *compatibilizing agent* to permit the formation of a homogeneous mixture of unmodified cellulose and the thermoplastic polymer, e.g., a cellulose-ethylene graft copolymer may be used to compatibilize cellulose and polyethylene."

The composition of the compatible mixture may be represented [2] as



$$\left(\begin{array}{c} A \\ A\text{———}x\text{———}B \\ B \end{array}\right) \qquad (5)$$

where A—x—B is a "compatibilizing agent" and compatibilizes Polymers A and B. However [1]

> "A graft copolymer containing polyhydric polymer segments and thermoplastic polymer segments may be used as a *compatibilizing agent* for a different polyhydric polymer and/or a different thermoplastic polymer, e.g., a cellulose-ethyl acrylate graft copolymer may be used to compatibilize starch and poly(vinyl chloride) due to the similarity of the solubility parameters of the segments of the graft copolymer with the homopolymers being compatibilized."

Block copolymers are generally more effective than graft copolymers as compatibilizing agents, particularly when numerous branches are located on a single backbone in the latter and restrict accessibility to the backbone segment of the copolymer. Nevertheless, the terms block and graft copolymers are frequently used interchangeably in discussions of compatibilizing agents since both contain segmental structures.

In view of the compatibilization of segments of copolymers with homopolymers of appropriate solubility parameter, an A—x—B block or graft copolymer may serve as a compatibilizing agent for various mixtures, including those containing polymers having structures different from those present in the compatibilizing copolymer.

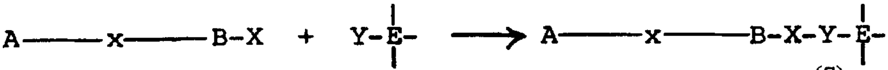
The nature of the bonding in the A—x—B compatibilizing agent is not restricted to covalent bonds but also includes ionic bonds, hydrogen bonds, and donor-acceptor bonds. The only limitation is that the bond remains intact during exposure to the conditions under which mixing with the incom-

$$\begin{array}{rcccl}
 & A & \text{—x—} & B & \\
 & A & + & B & \\
 & A & + & C & \delta C = \delta B \\
\delta D = \delta A & D & + & B & \\
 & D & + & C &
\end{array} \qquad (6)$$

patible polymers is conducted to promote compatibilization. The compatibilizing agent may be preformed and added to the mixture undergoing compatibilization or may be generated *in situ*.

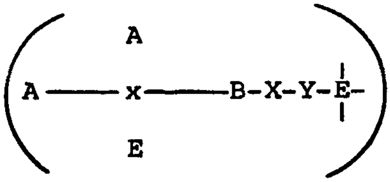
The minimum molecular weight of the segments in a compatibilizing agent may vary from that of a single reactive functionality to that of an oligomer or higher molecular weight polymer. However, a copolymer having a segment with a molecular weight above 100 000 is generally a poor compatibilizing agent since intra- and intermolecular interactions, such as chain entanglements, may reduce the accessibility of such a segment to the homopolymer whose compatibilization is desired.

Since compatibility is promoted by the presence of functional groups which undergo interaction, a segment of a compatibilizing agent containing reactive functionality (X) may compatibilize a polymer having a different structure as well as a different solubility parameter but having a coreactive functionality (Y), e.g.,



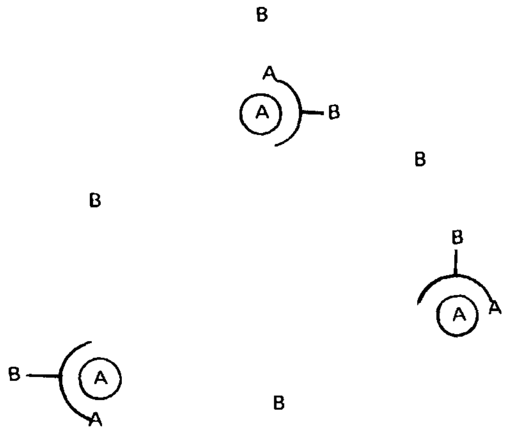
$$A\text{———x———}B\text{–}X + Y\text{–}\overset{|}{\underset{|}{E}}\text{–} \longrightarrow A\text{———x———}B\text{–}X\text{–}Y\text{–}\overset{|}{\underset{|}{E}}\text{–} \qquad (7)$$

The resultant graft or block copolymer, prepared *in situ* from a functionalized polymer containing pendent or terminal functionality is a compatibilizing agent for unfunctionalized A and E polymers.



$$\left(\begin{array}{c} A \\ A\text{———x———}B\text{–}X\text{–}Y\text{–}\overset{|}{\underset{|}{E}}\text{–} \\ E \end{array} \right) \qquad (8)$$

The compatibilizing agent functions as an organic surfactant in that it is located at the interface of two incompatible polymer phases, where it reduces interfacial tension and promotes adhesion between phases. It also promotes the formation and controls the particle size of the dispersed phase in a blend produced in the melt.



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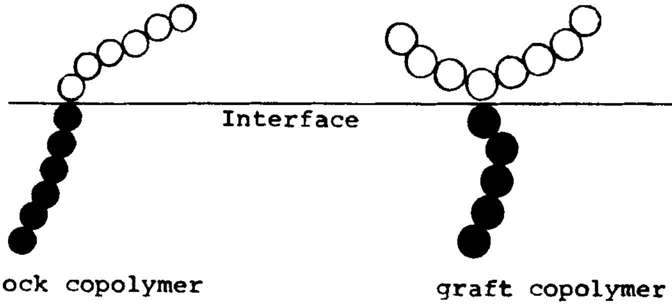


(10)

Analogous to the case of low molecular weight surfactants that are localized at the interface of oil and water phases in a stable emulsion, the effective concentration of compatibilizing agent or polymeric surfactant may be as little as 0.1% and is generally less than 5%, depending upon the desired particle size of the dispersed phase in a melt-blended or fabricated product.

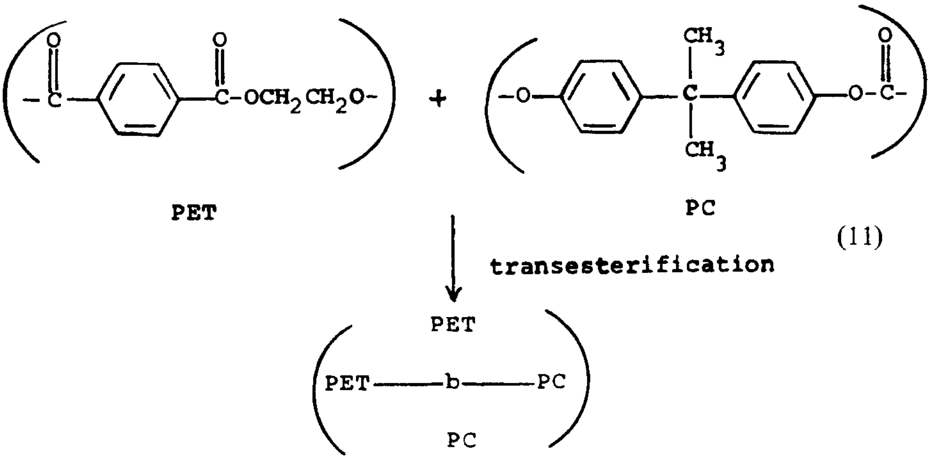
The *in-situ* formation of a compatibilizing agent at the interface of two incompatible polymers results in the generation of an adhesive bond between layers formed by extrusion lamination. The use of an adhesive layer or tie-coat permits the production of multilayer laminates possessing the desirable properties of several incompatible polymers. This concept is widely used in multilayer barrier packaging, where each layer has a specific function.

The term "blending" describes the process, generally conducted in the melt in mixing or extrusion equipment, in which two polymers are compatibilized to form a polyblend. The term "alloy" has been used in some cases to indicate that a chemical reaction between functional groups on the polymers results in covalent bond formation. However, the effectiveness of hydrogen and ionic bonding in promoting the formation of a compatibilizing agent in a blend of polymers indicates that this is a distinction without a difference.

POLYBLENDS AND ALLOYS

The role of compatibilizing agents in forming or stabilizing compositions containing otherwise incompatible polymers may be readily seen by reference to specific compositions in the technical and patent literature.

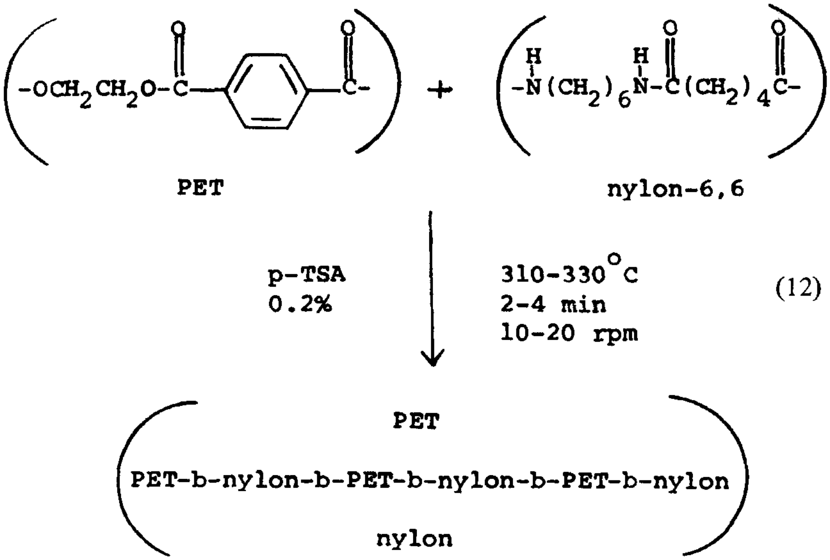
The *in-situ* formation of a block copolymer compatibilizing agent through an ester-ester interchange reaction is probably responsible for the observed compatibility of poly(ethylene terephthalate) (PET) and bisphenol polycarbonate (PC) [6, 7].



(11)

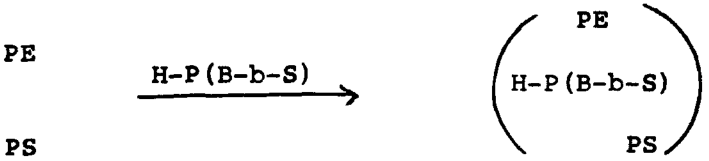
A PET-*b*-nylon 66 block copolymer is formed *in situ* through an ester-amide interchange reaction and compatibilizes a PET-nylon polyblend [8]. The compatibilizing agent is probably a multiblock copolymer, as shown.

Polyethylene (PE) and polystyrene (PS) are incompatible in admixture. However, the presence of a preformed hydrogenated butadiene-styrene



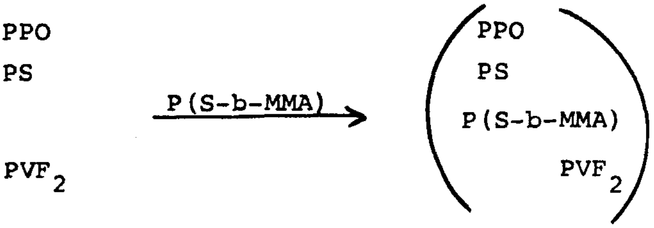
(12)

block copolymer, having the structure of a PE-*b*-PS copolymer, serves to compatibilize the incompatible PE and PS [9].



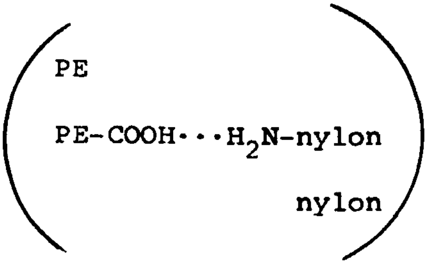
(13)

Polystyrene (PS) and poly(2,6-dimethylphenylene ether) (PPO) are compatible in admixture. However, neither PS nor PPO is compatible with poly(vinylidene fluoride) (PVF_2). On the other hand, PVF_2 is compatible with PMMA. A compatible multicomponent composition is produced by adding a preformed styrene-methyl methacrylate block copolymer [10].



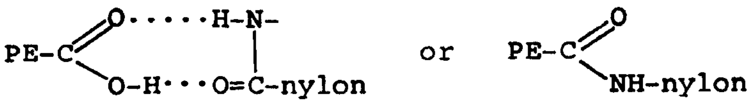
(14)

Mixing of PE and nylon 6 or nylon 66 with high shear was used to produce a polyblend having improved impact strength. The addition of a carbyxylated PE [11] or an ethylene-methacrylic acid random copolymer poly(E-*co*-MAA) [12] to a PE-nylon mixture resulted in a blend which produced shaped articles with resistance to permeation by organic fluids. The use of an ionomer, specifically a poly(E-*co*-MAA) copolymer partially neutralized with metal ions, yielded PE-nylon polyblends with decreased permeability to gases [13, 14].



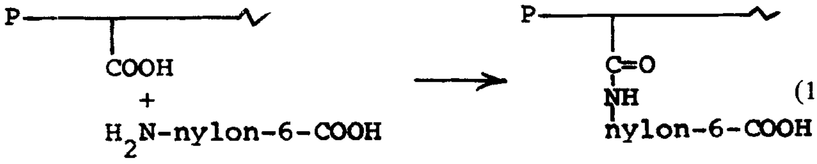
$$\left(\begin{array}{l} \mathrm{PE} \\ \mathrm{PE{-}COOH}\cdots\mathrm{H_2N{-}nylon} \\ \hfill \mathrm{nylon} \end{array}\right) \qquad (15)$$

The compatibilizing agent generated by interaction of the carboxyl-containing PE and nylon may result from hydrogen bonding or covalent bond formation.



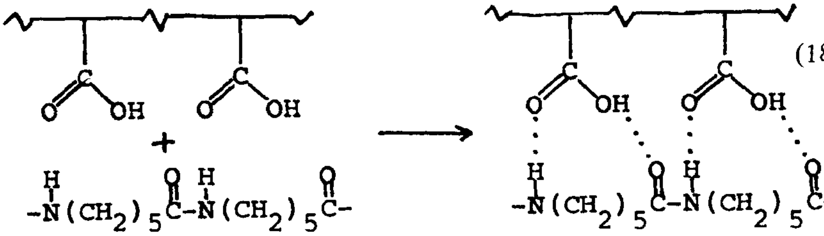
(16)

The interaction of a carboxyl group and the terminal amine group on nylon 6 was exploited in the preparation of an impact-modified nylon for blow molding applications. The carboxyl groups were present in an "acrylic modifier" such as a random copolymer of butyl acrylate, butylene glycol dimethacrylate, MMA, and acrylic acid (AA) or a graft copolymer prepared by grafting a mixture of MMA and methacrylic acid (MAA) onto a random poly(B-*co*-S) elastomer [15].



(17)

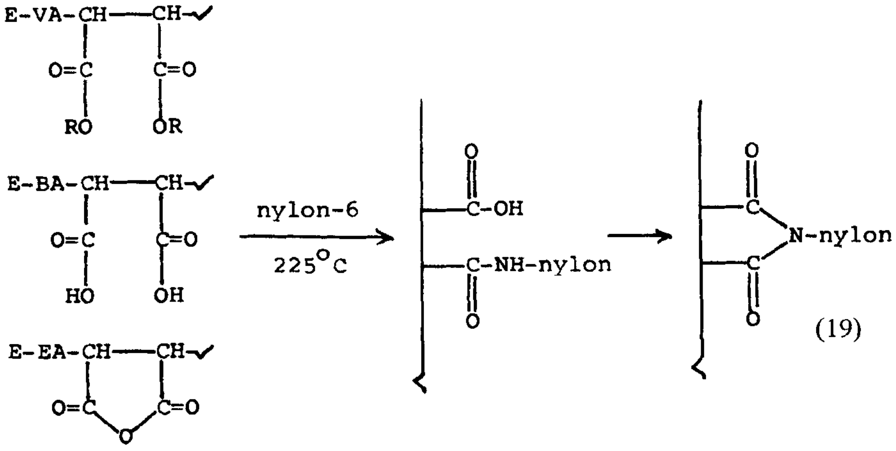
Since the indicated covalent bond formation probably occurs to only a limited extent, hydrogen-bonding interaction may play a dominant role in the carboxyl-containing "acrylic modifier"-polyamide interaction.



(18)

In contrast to the carboxylic acid-amine interaction, the reaction of a carboxylic acid anhydride with an amine readily results in covalent bond formation. The initially formed amic acid undergoes ring closure to a thermally stable imide at the elevated temperatures in an extruder.

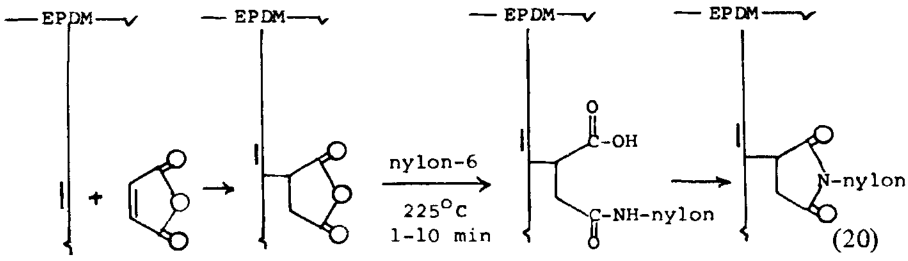
Copolymers containing maleic acid, maleic half ester, and maleic diester functionality undergo ring closure at elevated temperatures to form maleic anhydride (MAH) units. Elastomeric copolymers of ethylene with vinyl acetate (VAc), butyl acrylate, or ethyl acrylate, containing MAH or the acid or ester derivatives of MAH, react with nylon 6 in an extruder to yield an elastomer-nylon graft copolymer [16].



(19)

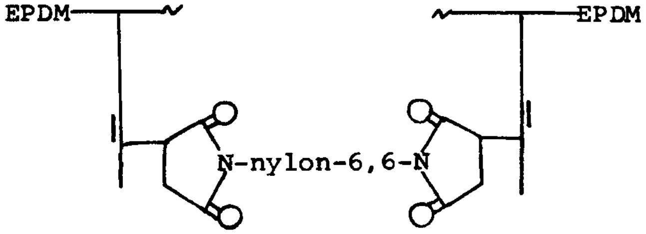
An MAH-containing ethylene-propylene-diene terpolymer (EPDM), prepared by the "ene" reaction of MAH and the unsaturation in EPDM at elevated temperatures, on admixture in an extruder with nylon 6 forms a rubber-modified nylon [16].

Since nylon 6 contains one terminal amine group, the product is a graft copolymer, free of crosslinking. The reaction of an MAH-containing elastomer, i.e., EPDM-MAH, with nylon 66 which possesses two terminal amine groups,



(20)

results in gel formation due to crosslinking. The product, commercially available from Du Pont as Zytel ST, is a "super-tough" nylon with greatly improved impact resistance. The compatibilizing agent is a nylon-crosslinked EPDM which contributes the impact resistance to the compatibilized blend of EPDM and nylon [17].

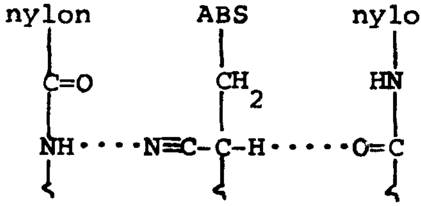


(21)

$$\left(\begin{array}{l}\text{EPDM}\\ \text{EPDM-MAH-nylon-MAH-EPDM}\\ \qquad\qquad \text{nylon}\end{array}\right) \qquad (22)$$

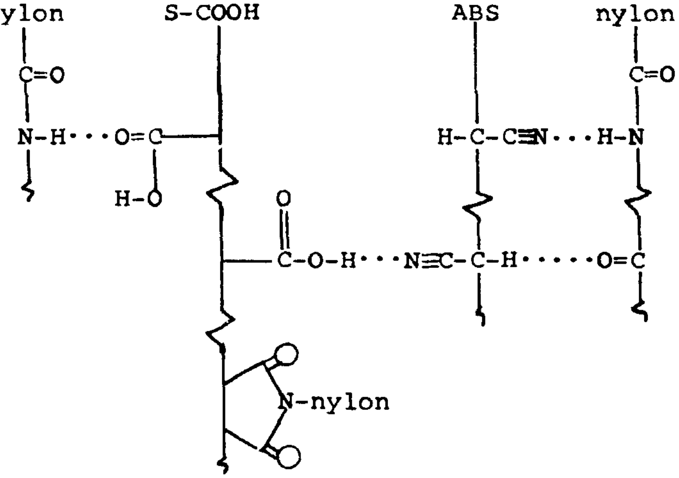
Poly(butadiene-*g*-[styrene-*co*-acrylonitrile]) (ABS) imparts its impact resistance to nylon in blends with it [18]. The compatibilizing interaction may be hydrogen bonding between the amide NH and the CN group in acrylonitrile (AN) or between the amide C=O and the α-methylene group in AN.

Improved impact resistance results from the addition of a carboxyl-containing polymer, e.g., poly(St-*co*-AA) or poly(St-*co*-AA-*co*-MAH), to the ABS-nylon presumably results from the presence of the carboxyl-containing polymer. Hydrogen bonding between the carboxyl group and the amide and/or



(23)

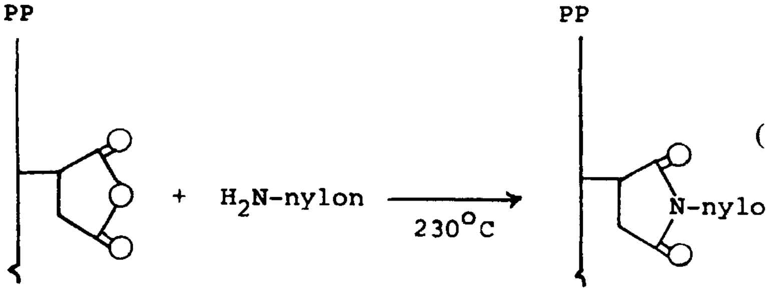
the α-methylene group of the AN is accompanied by reaction between the terminal amine group on the nylon and the anhydride, when present.



(24)

The amine-anhydride reaction promotes compatibilization of nylon 6 with polypropylene (PP) containing MAH-grafted PP [20]. The resultant polyblend shows improved water resistance and mechanical properties compared with nylon alone. Glass fibers are readily dispersed in the blend and increase modulus while undergoing interaction with the amide and anhydride groups.

The amine-anhydride reaction has been used in the preparation of an oil-resistant thermoplastic elastomer, commercially available as Geolast from the Monsanto Co. [21]. Conventional thermoplastic olefinic elastomers consist of blends of PP and EPDM, in which the EPDM is fully, partially, or not at all crosslinked. By virtue of their polyolefinic compositions, these materials are not oil resistant. In contrast, poly(butadiene-*co*-acrylonitrile) rubber (NBR) has oil resistance but can not be processed as a thermoplastic resin. By melt blending incompatible PP and NBR with PP-*g*-MAH and an amine-terminated



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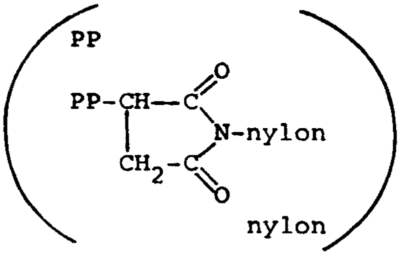
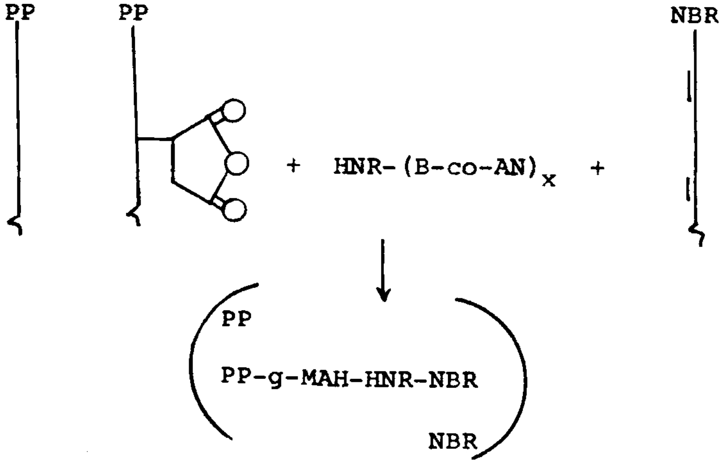


(26)

liquid NBR, the resultant compatibilized thermoplastic elastomer, after the NBR is vulcanized with a phenolic curing agent in the presence of a catalyst such as $SnCl_2$, is oil resistant.



(27)

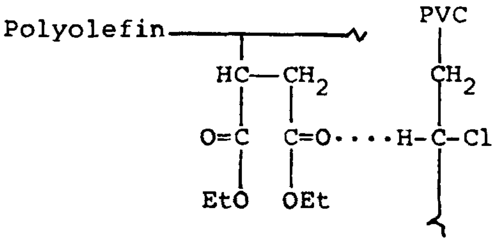
The concentration of amine-terminated NBR in the initial mixture is less than 2% and that of PP-*g*-MAH is less than 0.5%; higher concentrations have no effect on the resistance properties.

Hydrogen bonding between carboxyl groups and carbonyl groups undoubtedly plays a role in the preparation of "toughened" polyester by the melt blending of thermoplastic polyesters, such as poly(butylene terephthalate) (PBT), PET, and PC with acid- or anhydride-containing elastomers, such as poly(E-*co*-MA-*co*-monomethyl maleate), poly(E-*co*-isobutyl acrylate-*co*-MAA) ionomer, EPDM-*g*-MAH, and EPDM-*g*-fumaric acid [22].

High heat distortion temperatures and improved impact strength are obtained in interlocking network structures resulting from melt blending thermoplastic polyesters, such as PBT, polypivalolactone, or polycaprolactone, with dissimilar engineering thermoplastic resins such as polyamides, polyolefins, thermoplastic polyurethanes, halogenated thermoplastics, or nitrile barrier resins, in the presence of a hydrogenated triblock poly(St-*co*-B-*co*-St) copolymer which has a poly(St-*co*-EB-*co*-St) structure [23].

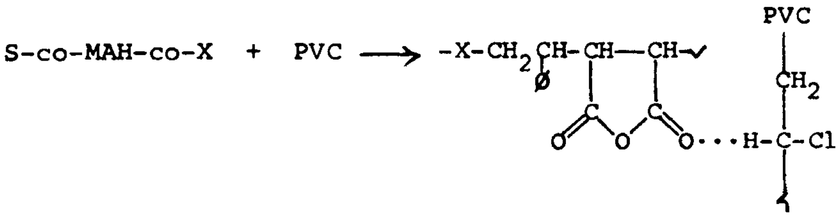
Significantly improved properties are noted in polyblends containing thermoplastic polymers and St-B-St triblock copolymers with as little as 2% MAH grafted on by the "ene" reaction in the case of the unhydrogenated copolymer or in the presence of a peroxide catalyst in the case of the hydrogenated copolymer. The anhydride-modified triblock copolymers are alloyed with polar thermoplastic polymers such as polyamides, polyesters, polyurethanes, polyacetals, polysulfones, polycarbonates, polyphenylene ethers, polyphenylene sulfides, nitrile polymers, ionomers, vinyl alcohol copolymers, e.g., E-VOH, and vinyl ester copolymers, e.g., E-VA [24]. The compatibilizing action is undoubtedly hydrogen bonding between the MAH carbonyl or carboxyl groups and the polar groups in the thermoplastic polymers.

Hydrogen bonding between the carbonyl groups in polyolefin graft copolymers containing diethyl maleate and the tertiary hydrogen in PVC results in the compatibilization of PVC and polyolefins, such as PE, PP, and EP [25, 26].



(28)

A similar interaction in the admixture of PVC and an St-MAH-X terpolymer has been claimed to increase the heat distortion temperature of PVC. The terpolymer, offered by Monsanto under the name Cadon 330, contains 25% MAH and an unspecified termonomer, probably an acrylic ester, and is promoted as an inexpensive "new engineering resin" [27].



(29)

An aromatic polycarbonate resin (PC) forms a compatible blend with an ABS resin. The latter normally contains a poly(St-*co*-AN) copolymer as well as the poly(B-*g*-[St-*co*-AN]) [28]. The properties of the polyblend are improved on the addition of an acid-containing copolymer, i.e., poly(St-*co*-AA-*co*-MAH) [29]. Improvements are also noted when a poly(St-*co*-MMA-*co*-MAH) copolymer is present in a polyblend of PC and ABS or MBS, i.e., poly(B-*g*-[St-*co*-MMA]) [30].

(PC
ABS)

(PC
P(S-co-AA-co-MAH)
ABS)

(PC
P(S-co-MMA-co-MAH)
ABS or MBS)

(30)

An impact-resistant blend of PPO and PS containing a hydrogenated St-B-St triblock copolymer is compatibilized with nylon 6 by the presence of a sulfonated PS ionomer.

(PPO
PS
P(S-b-E-B-b-S)
$PS-SO_3^-$ Zn^{++}
H_2N-nylon-6)

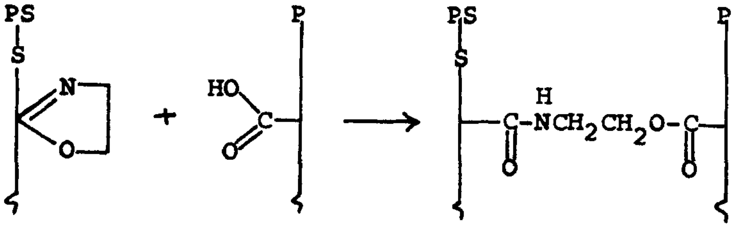
(31)

The styrene segments in the PS, the hydrogenated triblock copolymer, and the PS ionomer are compatible with each other and with the PPO, while the sulfonic acid moiety reacts with the amine group and compatibilizes the nylon. Analogous results may be obtained by the use of carboxyl-containing copolymers, e.g., poly(St-*co*-MAH), poly(St-*co*-AA), or carboxyl-containing or sulfonic acid-containing PPO.

Ionic bonds serve to compatibilize a blend of PPO and PS with a sulfonated EPDM ionomer. This is accomplished by the presence of a sulfonated PS ionomer which is compatible with the PPO and PS. Processability is improved, with retention of the properties of the polyblend, by the presence of plasticizers, e.g., triphenyl phosphate, which is a plasticizer for the PPO-PS blend, and zinc stearate, which is a plasticizer for the ionomers [31].

$$\left(\begin{array}{ll} \text{PPO} & \\ \text{PS} & (\phi\text{O})_3\text{PO} \\ \text{PS-SO}_3^-\ \ \text{Zn}^{++} & \text{Zn stearate} \\ \text{EPDM-SO}_3^-\ \ \text{Zn}^{++} & \end{array}\right) \qquad (32)$$

A styrene copolymer containing 1% oxazoline groups forms compatible polyblends with polymers containing coreactive functionality, such as carboxyl and anhydride groups as well as epoxy and amine functionality [32]. Thus, polyolefins are compatibilized with the "reactive polystyrene" (RPS) by the presence of functionalized polyolefin.



(33)

RECAPITULATION

The presence of a compatibilizing agent permits the blending of otherwise incompatible polymers to yield polyblends or alloys with unique properties, generally not attainable from the individual components. Block and graft copolymers possessing segments with chemical structures or solubility parameters which are similar to those of the polymers being blended are effective compatibilizing agents. The latter may be preformed and added to the mixture of polymers undergoing compatibilization or may be generated *in situ* by reaction between coreactive functional groups on the polymers. The bonding between the segments in the compatibilizing agent, whether preformed or generated *in situ,* may be covalent, ionic, or hydrogen bonding or donor-acceptor interaction. The compatibilizing agent acts as a polymeric surfactant, promoting interfacial adhesion between the dispersed and matrix polymer phases in a polyblend or alloy or between the layers in a laminate prepared by extrusion or coextrusion.

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