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Polyblend Compatibilization

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After an introduction regarding polymer blends, this review shows how the enhancement of the degree of compatibility between the constituent components of a polyblend can be made by using different reactive or additive compatibilizers. It also discusses how their presence affects the morphology and the properties of the polyblends. The compatibilization strategy is focusing on applications such as the recycling of post-consumer plastics without sorting, production of multi-layer films, packaging products, as well as the development of new materials based on plastic/plastic, elastomer/elastomer or plastic/elastomer blends.

Keywords polyblend, compatibilizer, morphology, properties

Introduction

Over the past decades, the need for new materials have forced researchers in academia and industry to consider polymer blend (alloy) systems and blending polymers, which is now one of the main topics of international polymer research. There are a number of reasons why researchers should have turned their attention to blends. For example, it has been shown that gaps in thermoplastics technology can be filled by carefully combining available polymers, that means, the use of blending. The process permits the combination of the attractive features of several materials into only one, to improve deficient characteristics of a particular polymer, and to reduce the price of an end product.

The growing commercial availability of polymer blends enlarges the diverse range of alternatives now being provided by the overall family of composites.

A polymer blend is a physical mixture of two or more polymers. The use of blends of two polymers eliminates problems like migration of one of them from the mixture, the second polymer being an additive with high molecular weight (MW), that means with high viscosity. Such blends can, in principle, offer a wide variety of morphologies and properties by adjustment of the nature and amount of polymers in the mixture; both these characteristics strongly depend also on their miscibility. Historically, polyblends were developed to improve the impact strength of rigid polymers (1).

More recently, polymer blending has also been employed to enable the reuse of recycled industrial and/or municipal plastic waste (2–4).

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Beside the binary systems, ternary blends are gaining importance (5, 6).

In blending different polymers, one may encounter two extreme situations: a gross phase separation, or a homogeneous mixture at the molecular level. However, the reality is more complex, and many intermediate situations can occur: increasing separation at the intermolecular level only, formation of co-continuous morphology, dephasing into dispersed heterophase morphologies of increasing phase size (7).

The formation of two phase systems is not necessarily an unfavorable event since many useful properties, characteristic of a single phase, may be preserved in the blend while other properties may be averaged according to the blend composition. Proper control of overall blend morphology and good adhesion between the phases are in any case required in order to achieve good mechanical properties. For instance, the toughness properties of immiscible blends whose components form separate phases often depend on the dimensions of the dispersed phase and on the degree of interfacial adhesion between phases. These issues are largely controlled by melt processing conditions and by the thermodynamic affinity of the components of the polyblend.

There seems to be a general agreement on the fact that there is no compatibility between two polymers when they are not able to develop either weak or strong specific interactions. The formation of a polymer blend seems to be due to a balance between a free volume contribution (always unfavorable) and an interaction term (potentially favorable). The later one, in turn is a balance between self-association (P-P, P*-P*) and hetero-association (P-P*). Much work has been done trying to predict whether P and P* polymers are compatible prior blending, mainly using the value of solubility parameter, IR spectroscopy, electrical mechanical techniques (8), or molecular mechanic calculations (9).

The specific interactions between polymers are essentially electrostatic: dipole-dipole, hydrogen bond, charge transfer (10).

In general, polymers scarcely mix with one another, because the combinational entropy (thermodynamic reasons) between polymers is considerably low. As miscibility of polymers strongly depends on polymer-polymer interaction, a slight difference in conformation of polymer chain sometime affects this process (11). Blending does not result in many cases in a stable finely dispersed distribution of a polymer in the other one. Such type of distribution is necessary to get a sufficient interaction between polymers, which results in the formation of interfaces, i.e., in the formation of at least one new polymer phase (12).

It is well established that parameters like control of blend morphology and phases adhesion are strongly dependent on the presence in the blend of appropriate block or graft copolymers having chain segments identical or similar to the homopolymers to be mixed.

All the phase behavior displayed by polyblends can be found in mixtures of small molecules, but in different ways, taking in account the differences between micro and macromolecules. For these reasons, different terms like miscibility and compatibility became necessary for polymer blends studies.

Miscibility is the same as thermodynamic solubility; that is two polymers are miscible in each other if the free energy of mixing (ΔG_{mix}) them is negative. Miscibility has a precise definition that allows it to be measured accurately for the determination of phase diagram of a system. As such, this is a function of the micro structure of the polymers (e.g., M.W., polydispersity, tacticity, presence of comonomer), and thermodynamic variables such as temperature, pressure and blend composition. Two polymers, which are immiscible at a 50/50 ratio can be miscible at a 95/5 ratio under the same conditions.

While miscibility has a strict thermodynamic meaning, compatibility is defined in operational terms. A blend can be more or less compatible if it is closer or further from miscibility. Greater or lesser degrees of compatibility measured will depend on the technique used and the scale at which it probes the blend structure. It is a relative term in the sense that the phase size of the blend will change with the time depending on the kinetics of phase separation. Always is necessary to specify the method used to determine compatibility (glass transition temperature, T_g , microscopy, rheology, light scattering, etc.).

A completely compatible system is thus a miscible one. The compatibility of two polymers of a particular blend can be related to how good a particular property is. How the degree of compatibility of an immiscible system is measured or determined depends to some extent on why some one is interested in the blend, often in terms of what application it can see. There are ways to change properties without affecting the phase separated domain size and growth, which are system responses which reflect the thermodynamic driving forces for phase separation.

C. Koning et al., the authors of strategies for compatibilization of polymer blends (13), classify different polymer blends in:

- Completely miscible blends
- Partially miscible blends (compatible)
- Immiscible blends

In miscible blends, for which $\Delta H_{mix} < 0$, due to specific interactions, homogeneity is observed at least at the nanometer scale, if not at the molecular level (example polystyrene and polypropylene oxide blend).

In the second group, part of one component is dissolved in the other and the blend exhibits a fine phase morphology and satisfactory properties. Both phases (one being rich in polymer P, and the other in polymer P* are homogeneous and have their own T_g . (Example: polycarbonate, PC, and acrylonitrile-butadiene-styrene terpolymer, ABS.)

The immiscible blends have a coarse phase morphology, the adhesion between phases is poor, so they cannot be used without a compatibilizer. Most of the polymer blends are immiscible.

As it was already mentioned, the formation of miscible polymer blends requires that the free energy of mixing,

$$\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix}$$

where ΔH_{mix} and ΔS_{mix} are the enthalpy and entropy changes, respectively, be negative and that $\{\delta^2 \Delta G_{mix} / \delta \varphi_i^2\}_{T,P} > 0$ where φ_i is a measure of the concentration of component i , and δ is the solubility parameter. Since the entropy of mixing high MW macromolecules is quite small, miscibility usually occurs when the enthalpy of mixing is negative (14–17).

The entropy of mixing is a function of the MW, decreasing rapidly toward zero as the degrees of polymerization of the components approach the values typically found in commercial polymers. The enthalpy of mixing, on the other hand, primarily depends on the energy change associated with changes in nearest neighbor contacts during mixing and is much less dependent on MW (18).

Polymer miscibility is usually studied by direct observation, either visual or microscopic, of a blend specimen, usually in the form of a thin film, or by observation of its T_g and crystalline melting points (T_m). Miscible amorphous polymers form transparent

films and exhibit no heterogeneity in a phase contrast microscope or in an electronic microscope. Furthermore, immiscible fully amorphous polymers form transparent films when polymers have the same refractive index, or when the refractive indices are different, they occasionally form two-layered films that appeared transparent when the blend solution is evaporated. Miscible polymers involve thermodynamic solubility and should have one phase and only a single T_g . Heterogeneous, immiscible blends are phase separated, exhibiting the T_g s and/or T_m s of the components. If there is partial miscibility, the T_g s move closer together compared with the individual components. If, on the other hand, there is complete immiscibility, then the T_g s of the individual phases are identical to the T_g s of the components. If specific interactions occur between the two components P and P*, then to a rough approximation, T_{gPP^*} in the homogeneous blend is the weighed average of T_{gP} and T_{gP^*} .

A precise estimated value of T_g of a blend is obtained with the aid of the Flory-Fox equation:

$$\frac{1}{T_{gPP^*}} = \frac{W_P}{T_{gP}} + \frac{W_{P^*}}{T_{gP^*}}$$

where W_P and W_{P^*} are the weight fractions of the blend components P and P* (16).

The highest degree of compatibility (that is miscibility) does not always mean the best engineering properties. In some cases, such as impact strength of plastics, some amount of phase separation is necessary to obtain the desired values. The goal is to achieve a controlled level of phase separation. However, in most cases, due to the preponderance of immiscibility, the need is to increase compatibility. The mechanism and dynamics of phase separation in polyblends has long been a research subject in an attempt to obtain high performance materials by controlling morphology.

Numerous studies have proved that the presence of any kind of interactions between the polyblend components such as hydrogen bonding, ion-ion pairing, electrono-donor, electrono-acceptor complexation, etc. produces a favorable mixing enthalpy and hence can lead to complete miscibility. Such findings provide a practical guide for studying miscibility enhancement in polyblends, i.e., by chemical modification, copolymerization, introduction of groups able to form specific interactions (19–21). The energy of interaction between polymer units increases in the order: van der Waals interactions < dipole-dipole interactions < electron donor-acceptor complex formation = H bonding = ion-dipole interactions < ion-ion interactions (22).

Compatibilization

The general incompatibility of polymers prevents preparation of useful blends, but adding to the system of a compatibilizing agent permits the blending of otherwise incompatible polymers to yield compositions with unique properties generally not attainable from either of the mixture components. The presence of such an additive influences the properties of polymer melts and solutions.

A variety of additives can be used to promote miscibility by reducing the interfacial tension. Reactive compatibilizers chemically react with blend components and are, therefore, effective for many systems. Nonreactive compatibilizers are typically block and graft copolymers of the partners and are more specific in their action. They can be useful in improving interfacial coupling (23).

Proper interfacial tension leads to a phase size small enough to allow the material to be considered as macroscopically homogeneous. A strong interphase adhesion can assimilate stresses and strains without disruption of the established morphology (24).

In fact, the addition of a small amount of a third ingredient to a binary blend stabilizes the phase morphology. A block or a graft copolymer, as mentioned before, which works as an emulsifier is very effective in reducing the interfacial tension and improving the interfacial adhesion between macromolecules, and hence, the mechanical properties of the polyblend product. In some cases can be effective the addition of higher amounts of a core-shell copolymer that behaves like a multi-purpose compatibilizer-cum-impact modifier (25).

Since polymer properties are derived from finite length segments of a particular structure, random copolymers do not compatibilize homopolymers, and in many cases, copolymers based on one ratio of co-monomers are not compatible with copolymers based on a different ratio of the same co-monomers. Block copolymers provide a stronger interfacial adhesion. For this reason, such copolymers are most commonly used. Symmetric reactively formed block copolymer with higher MW is desirable mostly for reactive compatibilization (2, 26). Many studies on the compatibilization of polymers, however, use the graft copolymer as the compatibilizing agent. Similar to block copolymers, the MW of a graft copolymer plays also a fundamental role; if it too short, the copolymer is not efficient as a compatibilizing agent. The number and length of branches play an important role in the compatibilization process. Rather than adding, it is often preferable to obtain a graft copolymer *in situ* by means of a chemical reaction (2).

A. Ajji and L. A. Utracki (27) show that the objectives of the compatibilization process are to:

1. Reduce the interfacial tension, facilitating the dispersion,
2. Stabilize morphology against high stress and strain processing, and
3. Enhance adhesion between the phases in the solid state, thus improving the properties of the polyblend.

The ability (or efficiency) of a compatibilizer to decrease the domain size of the dispersed phase in blends depends on:

- the interaction parameter (or interfacial tension) between the components
- the difference in viscosity (MW) or elasticity between the components
- the amount and type of compatibilizer
- the processing parameters such as temperature, shear rate, etc.

The polymers that affect the miscibility generally do not act as interfacial agents, but rather operate by modifying the phases properties.

Ionomers have served as compatibilizers in many systems, ionic or other strong physico-chemical interactions generated across the interface having effected the compatibilization (28).

Reactive Compatibilization

N. C. Liu and W. E. Baker (29) discuss several methods for compatibilization, such as: introduction of non reactive graft or block copolymers, non-bonding specific interactions, low MW coupling agents and reactive polymers.

Non-bonding specific interactions (hydrogen bonding, ion-dipole, dipole-dipole, donor acceptor and π -electron interactions) are useful for enhancing the compatibility of polymer blends. Besides the additive compatibilization (incorporation of a polymer, a lubricant or another compatibilizer agent), reactive blending is used.

The formation "*in situ*" of a block or graft copolymer by blending suitable functionalized polymers is known as reactive compatibilization. M. Moan et al. (30) studied experimentally the effects of such a compatibilization on morphological and rheological properties of blends of a polyamide (PA) dispersed in a polyethylene, (PE). The location of the copolymer at the interface can lead to a reduction of the interfacial tension and enhancement of the adhesion. The reactive compatibilization is realized through covalent or ionic bonding during the melt blending of suitably functionalized polymers. B. Majumdar and D. R. Paul (31) show that the focus here is on block or graft copolymers generated *in situ* during the process of melt mixing, through a reaction between functional groups available on macromolecules and the changes in blend morphology and properties. Reactive compatibilization is used by many researchers for different polyblend systems (32–40). It is important to note that the generation of morphology in multiphase polymeric systems through chemical reactions is a dynamic process.

The study of polypropylene (PP) and polyhydroxyaminoethers (PHAE) blends of different compositions was done by R. Zacur et al. (32). Maleic anhydride grafted PP, MA-g-PP was found to be an effective compatibilizer.

The reactive extrusion of an epoxy compatibilized blend of poly(ethylene terephthalate) (PET) and a thermotropic liquid crystal polymer (LCP) was studied by F. Qiao et al. (33).

F. Pazzagli and M. Pracella (4) report on a study of melt free radical grafting of high density polyethylene (HDPE) both virgin and recycled with glycidyl methacrylate monomer (GMA) and reactive mixing with recycled PET. The effect of a grafting procedure, radical initiator and co-monomer content on the grafting degree of HDPE and properties of PET/HDPE-g-GMA blends was investigated. The results point out the great potential of melt grafting functionalization and reactive mixing for processing and upgrading recycled polymers.

G.H. Hu et al. (34) report the feasibility of the process called *in situ* polymerization and *in situ* compatibilization of PP/PA6 polyblends. These blends were prepared by activated anionic polymerization of ϵ -caprolactam in the presence of PP in a batch mixer and a twin screw extruder.

Studies on one-step *in situ* compatibilization of recycled PET and recycled PA were also done in view of process optimization (35).

A random terpolymer based on MA, acrylic ester and ethylene(E) has been used for an immiscible system based on PE and PA. The reaction between the anhydride and the end group of polyamide occurs during mixing and the *in situ* formed graft copolymer acts as a compatibilizer. The terpolymer can be considered compatible with PE because of its high content (89.5%) of ethylene (30).

PA6/ABS blends were obtained by W. Rongwei et al. (41) using a SAM which contained 6wt% MA, 19 wt% AN, and a graft ABS powder containing 68% PB rubber. The blends with 50/50–70/30 ratios of PA6 to ABS at about 4% SAM exhibited good toughness at room temperature and excellent low temperature toughness. These can be evidently attributed to the fact that the *in-situ* reactive compatibilization makes the interfacial tension reduced and the size of ABS domains more uniform through the blends.

The effectiveness of some thermoplastic elastomers grafted with MA or with glycidyl methacrylate (GMA) as compatibilizer precursors for blends based on LDPE and PA6 has been studied by S. Filippi et al. (42). The obtained data confirm that the MA functional groups possess considerably higher efficiency for the reactive compatibilization of LDPE/PA6 blends, than those of the ethylene-acrylic acid and ethylene-GMA copolymers.

D. Hlavata et al. (36) studied the reactive compatibilization using liquid polybutadiene (PB) and diallyl peroxides in the case of low density polyethylene LDPE/PS (4/1) blends and the commingled waste of composition similar to these blends. The influence of lubricants on the structure and toughness of these blends was determined. The reactive compatibilization had the same effect in both types of the blends, but its effect is enhanced by addition of lubricants.

An *in situ* compatibilizer of poly(styrene-ran-glycidyl methacrylate) (PS-GMA) was used by J. K. Kim et al. (37) to study the effect of blend compositions on the morphology of two different systems, i.e., poly(ethylene-ran-acrylic acid) (PE-AA) and PS blend, and poly(butylene terephthalate) (PBT) and PS blend. The domain size of the dispersed phase in the PE-AA/PS blend system was larger than that in the PBT/PS blend at the same blend composition when there is no PS-GMA in both blends.

M. T. Ramesan et al. (24) established that dichlorocarbene modified styrene-butadiene rubber (SBR) acts as a compatibilizer in the blends of SBR and acrylonitrile-butadiene rubber (NBR). The efficiency of compatibilization depends on the chlorine content of the compatibilizer and the blend ratio of the components. The compatibilizing efficiency increases as the SBR content in the blend decreases and also when chlorine content of the compatibilizer increases up to 25%.

Hydrogenated segmented poly[butadiene-block-((styrene-co-acrylonitrile)-block-butadiene)n] block copolymers, developed by the use of the polymeric iniferter technique, were tested for their compatibilizing capacities for 10/90 LDPE/PVC blends by E. Kroeze et al. (38). The acrylonitrile content of the SAN blocks of the block copolymers was as expected, found to be extremely important factor for their miscibility with the PVC phase.

Y. Haba and M. Narkis (43) obtained PVC/PS blends through a reactive extrusion-polymerization by the absorption of a solution of S, initiator and crosslinking agent in porous PVC particles, forming a dry-blend with a relatively high monomer content. The monomer contained in particles was polymerized in a twin-screw extruder in the melt state. After polymerization, no residual monomer was detected by GC. The transparency, fracture surface morphology, thermal stability, rheology, and static and dynamic mechanical properties of these blends were compared with the PVC/PS blends obtained by physical mixing and having the same compositions.

G. H. Hu et al. (39) showed that an efficient removal of residual monomers is very important for *in situ* compatibilization of immiscible polymer blends by one-step reactive extrusion. Two model blends were used for the study; PP/PBT and HDPE/PA6. A co-rotating intermeshing twin screw extruder was used to process these blends. GMA and MA were applied to functionalized PP and HDPE. Results showed that the elongation at break and impact strength of both blends were improved to a much greater extent with up-stream devolatilization compared with down-stream devolatilization.

D. A. Baker et al. (40) have used azide cross-linking agents that attach PP chains to PS chains in a PP/PS blend. The azide group upon heating loses nitrogen, to form highly reactive nitrene species that inserts to C-H bonds in the polymer chains. Thermal analysis indicated that PP was chemically attached to PS.

Reactive compatibilization of the ternary system PE/PP/PS was studied by I. Fortelny et al. (6). They found that PB/organic peroxide system can be used as a reactive compatibilizer, which somewhat increases the toughness of LDPE/high impact PS/HIPS blends, and that styrene-butadiene copolymer (SB) is an efficient compatibilizer for LDPE/PP blends with higher than 40wt% LDPE contents.

These authors also established that the compatibilization with liquid PB and an addition of SB copolymer, substantially enhances the toughness of LDPE/HDPE/PP/HIPS blends with composition similar to the municipal plastic waste.

The reactive compatibilization of ethylene-vinyl acetate copolymer (EVA)/NBR has been performed by B. G. Soares et al. (44) by using partially hydrolyzed EVA in combination with oxazoline-functionalized NBR. The presence of these compatibilizers in certain amounts resulted in a substantial improvement of tensile strength of EVA/NBR (50:50 wt%) vulcanized blends, with a little increase of the elongation at break.

The reactive compatibilization of ethylene-propylene-diene monomer EPDM/NBR blends was performed by M. G. Oliveira et al. (45) using mercapto modified copolymers such as: EPDMSH (mercapto modified EPDM), EVASH (mercapto modified EVA) in combination with oxazoline-functionalized NBR (NBR-ox). The best mechanical properties were obtained with the two EVASH- based compatibilizing systems, especially the one containing the co-reactive copolymers because of the reactivity of the oxazoline group. The presence of insoluble material in non-vulcanized blends suggested the reactive compatibilization.

Dynamically vulcanized PP/EPDM blends were treated by high intensity ultrasonic waves during extrusion by W. Feng and A. I. Isayev (46). These blends were compared with unvulcanized blends of the same polymers that were treated by ultrasound during extrusion and then dynamically vulcanized. The results obtained indicated that ultrasonic treatment induced the thermomechanical degradation causing enhanced molecular transport and chemical reactions at the interfaces, thus leading to *in situ* compatibilization, which is evident by the morphology and mechanical properties.

Solid state shear pulverization (SSSP) has been shown to achieve compatibilization of immiscible polymers by the *in situ* formation of block copolymers resulting from coupling of macroradicals made via low level scission (47).

L. Duxin et al. (48) studied the compatibilization of the PP/PA6 blend by using a PP solid-phase graft copolymer (gPP). This one improved efficiently the compatibility of PP/PA6 blend. Due to the reaction between the reactive groups of gPP and the NH₂ end groups of PA6, a PP-g-PA6 copolymer was formed as a compatibilizer in the vicinity of the interface during the extrusion melting of gPP and PA6. The compatibilized PP/PA6 blends possessed higher pseudoplasticity, melt viscosity and flow activation energy.

Compatibilizing Agents

For both virgin and regenerated incompatible polyolefin blends like LDPE/PP, S. Bertin and J. J. Robin (49) have used various compatibilizers like EP or EPDM block copolymers. All these block copolymers led to better properties than those obtained with the graft copolymers.

Hydroxyl functionalized copolymers were obtained by U. Hippi et al. (50) through the copolymerization of propylene with 10-undecen-1-ol and used as compatibilizer for PP/PA6 blend with a composition of 70/30. Enhanced adhesion between the blend components was observed in morphology and DMA studies. Although improvement in

toughness was not as pronounced as expected, there were indications that the hydroxylated copolymers prepared with metallocene catalysts could serve as a new type of compatibilizers.

B. J. Kim and J. L. White (51) applied lauryl lactam (LA)-caprolactone (CL) block copolymer P(LA-b-CL) as a compatibilizer for immiscible PVC blends with other various polymers. The obtained blends possess high thermal performance and toughness.

The compatibilization of PC with ABS was realized by W. Jeroen et al. (52) with core-shell particles. The sub-micron sized additive, consisting of a butadiene rubber based core and an acrylic based shell, position themselves on the PC/SAN interphase and act as a compatibilizer in such blends. Besides the compatibilization effect, the elastomeric core of the compatibilizer leads to a significant improvement of performance in terms of low temperature impact, impact after ageing and weld line strength.

S. H. Zhu et al. (53) studied the effect of NBR compatibilizer on the mechanical properties of PVC/SBR blends. A significant improvement of these properties was established when the blend was vulcanized in the presence of the compatibilizer due to a covulcanization between NBR and SBR. The fracture toughness of PVC/NBR/SBR (50/10/40) blends was characterized by the critical strain energy release rate G_c . In the case of some of such blends, an increase in sulfur concentration resulted in an important increase in G_c .

For a composite based on a copolymer of ϵ -caprolactam and AH salt with ultrathin PP fibers M. V. Tsebrenko et al. (54) have used the EVA copolymer and sodium oleate as a compatibilizer.

A. E. Zaikin et al. (55) established that the introduction of a filler into a blend of strongly dissimilar polymers like NBR and EP random copolymer should decrease their mutual solubility and hence, their mutual adhesion. Filler (aerosil) particles with strongly absorbed macromolecules of both polymers were formed at the interface.

The research done by C. Xu et al. (56) concluded that NBR can promote the phase dispersion of PVC and LDPE and their interfacial adhesion.

Chlorinated PE (CPE) was used by E. A. Eastwood and M. D. Dadmun (57) for the compatibilization of PVC and a polyolefin elastomer (POE). The interfacial adhesion force is optimum at 20% chlorine content of CPE and increases with its MW increase. The POE-CPE interaction was found to govern the ability of the CPE to compatibilize PVC with POE.

M. Singer et al. (58) obtained blends of a LCP with PC; a copolymer of methyl methacrylate (MMA) and dimethylglutarimide was used as compatibilizing agent. It was found that this compatibilizer yields a fibrillar blend morphology, probably due to reducing interfacial tension and adhering to the blend phases.

PE-g-LCP copolymers were used by F. P. La Mantia et al. (59) as compatibilizer for blends of PE and semi-rigid LCP. Such additives lead to an improvement of interfacial adhesion, both in melt and in solid state, as well as to a modest enhancement of the mechanical properties.

The compatibilization efficiency of two styrene-butadiene-styrene SBS triblock copolymers with short and long styrene blocks was studied in PS-PP blends of composition 20, 50, and 80% PS by D. Hlavata et al. (60). The supramolecular structure of the blends was determined by small-angle X-ray scattering, and the morphology was established with transmission (TEM) and scanning electron microscopy (SEM). The compatibilization efficiency of SBS copolymers in PS-PP blends depended on the conditions of blend mixing and processing and could not be predicted with the rules of equilibrium thermodynamics only.

Recently, the treatment of wood flour with a commercial paper wet-strength agent before mixing it with stearic anhydride treated PE, increased the modulus of rupture and the modulus of elasticity of the resulting wood-PE composites (61).

The role of the polymer architecture in the compatibilizing process of PS/PMMA blend was studied by E. A. Eastwood and M. D. Dadmun (62). Diblock, triblock, pentablock and heptablock copolymers of S and MMA with similar MW were compared to a random copolymer. Strengthening of the PS/PMMA interfaces was attributed to multiple interface crossings. Failure of the heptablock copolymer to give relatively strong interfaces was ascribed to the block lengths that are insufficient to entangle with and anchor in the homopolymer. Unlike data using random copolymer as interfacial modifiers, dependence of the fracture toughness on the copolymer composition was not observed for the multi-block copolymers studied. S centered and MMA centered multi-block copolymers of each type demonstrated similar results and the increasing lengths of blocks increased the interfacial adhesion. MWs of the blocks have to be high enough to obtain significant anchoring in the copolymers.

The effect of reactively formed block and graft copolymers on compatibilization of 70/30 wt/wt PA66/PS blends has been investigated by H. K. Jeon et al. (26) by observing morphology and measuring reaction conversion. Anhydride functional PSs were loaded up to 10 wt% in the blends. From the comparison of the PS particle sizes and the estimation of interfacial stability by observing micelles, it was found that the higher MW PS-An (phthalic anhydride terminal PS) is the most effective compatibilizer, showing finer morphology and higher interfacial stability. This implies that symmetric reactively formed block copolymer with higher MW is desirable for reactive compatibilization. Blends of PA66 with syndiotactic PS compatibilized with PS-An gave very similar morphology to the PA66/PS blends indicating that this conclusion also applies to PA66/syndiotactic PS blends.

A. Benderly et al. (63) found that the compatibilization process in the PP/PA6/glass fiber system seems to be significantly slower than the process of filler encapsulation. To prevent this encapsulation, the filler must be added to an already compatibilized polymer melt.

An ethylene-methacrylic acid copolymer partially neutralized with sodium (Na-EMAA) was successfully used by A. Lahor et al. (64) to compatibilize PA6 and LDPE. The phase morphology and thermal characteristics of these blends were investigated over a range of compositions.

At certain ratio between the polymers, dimethyl formamide (DMF) may lead to high miscibility of PVC and PMMA blend (65).

A completely different technique was applied by H. Li and E. Ruckenstein (66), consisting in a self-compatibilization, by mechanically mixing concentrated emulsions of two oligomers and then continued the polymerization process. Some compatibilizers are generated at the interphase. In particular, when a rigid polymer was blended in this way with a flexible one, tough materials were obtained. The effect of compatibilization can be evaluated from the impact strength, which constitutes a proper measure of toughness. The greater the amount of functional groups, the higher the impact strength. This occurs because of a higher content of functional groups resulting in a greater amount of gel, which is mostly composed of rigid and flexible chains. Such a combination inhibits the propagation of cracks in the matrix and hence increases the impact strength.

Epoxidized natural rubber (ENR) was used by P. I. Teh et al. (67) as a compatibilizer for nanocomposites based on natural rubber (NR) and an organophilic layered clay (organoclay).

Montmorillonite clay was added at different amounts to blends of NBR and SBR. Both the reinforcing and compatibilizing performance of this filler were investigated using rheometric measurements, physico-mechanical properties, SEM and DSC (68).

Effects on the Morphology and Properties

Control of the phase morphology during blend processing is a key issue for the production of new materials with improved properties compared with those of constitutive immiscible polymers. The shape, size, and spatial distribution of the phases result from a complex interplay between viscosity of the phases, interfacial characteristics, blend composition and processing parameters (13).

Polyblends often exhibit poor mechanical properties due to incompatibility resulting from the lower entropy of mixing of high MW polymers and the unfavorable enthalpic interaction between them. As a result, their morphology is characterized by phase separation.

The mechanism and dynamics of phase separation in polyblends have long been a research subject in an attempt to obtain high performance materials by controlling morphology (69).

Polyblend compatibilization is directly related to blend morphology and phase sizes (25).

The *in situ* compatibilization of polymer blends in the melt state was found to occur during ultrasonic assisted extrusion at high pressures and temperatures without use of any chemicals. The mechanical properties of ultrasonically treated blends were significantly improved as compared to the untreated blends. Following A. I. Isayev et al. (70), this new technique can be applied for plastic/elastomer blends to make thermoplastic elastomers or plastic/plastic and elastomer/elastomer blends and for making new copolymers from practically any pairs of existing polymers to achieve desirable chemical and physical properties.

H. Cartier and G-H. Hu paper (71) deals with the morphology development of *in situ* compatibilized polymer blends in a co-rotating twin-screw extruder. Using blends made of two semicrystalline polymers, PP and PA6 of different T_m , it has been found that the morphology of *in situ* compatibilized blends with MA modified PP (PPgMA) develops much faster than that of their mechanical counterparts. The finding that the morphology of the PP/PA6 reactive blend develops rapidly made it possible to produce compatibilized blends by one-step reactive extrusion that integrates the traditionally separated free radical grafting of MA onto PP and the compatibilization of PP/PA6 into a single more economical extrusion.

The dependence of the morphology development of physical and, as well as reactive compatibilization of PP/PA6 blends in a mixing zone of a co-rotating twin screw extruder on blend composition and screw rotational speed, was investigated by O. Franzheim et al. (72).

Melt blends of thermoplastic polyurethane (TPU) and poly(styrene-co-acrylonitrile) SAN, of various ratios were studied by S. N. Jaisankar and G. Radhakrishnan (73). Blend preparation was done by using a two-roll mill, and the compatibilization effect of S-MA on these incompatible polymers was studied. The morphology and the properties of the blends were investigated using SEM, DSC, Fourier transform infrared spectroscopy (FTIR) and mechanical testing. TPU/SAN blends 70:30 and 50:50 show better mechanical properties than other blend ratios. The addition of 5% S-MA improved the miscibility as evidenced by uniform phase (SEM micrographs) and thermal-mechanical properties of

the same blends via melting mixing were studied by M. Ulcnik-Knump and F. L. De Lucca (74). The compatibilizers used were; poly- ϵ -caprolacton (PCL), a mixture of PS-*b*-PCL and PS-*b*-PMMA, and a mixture of polyisoprene-*b*-PCL and PB-*b*-PMMA. The blends compatibilized with PCL showed superior properties over the other blends.

Morphology studies of PP/PS blends were performed by O. J. Danella, Jr. and S. Manrich (75) based on torque and viscosity characteristics. The processing was done in a twin-screw extruder, without and with a compatibilizing agent. SEM images of the PP/PS blends 80/20 wt% with no compatibilizing agent showed an increase in both areas and average diameter of the PS phase in PP/PS blends with similar torque ratio when compared to those with a larger torque ratio. Also, three different copolymers, PP-*g*-MA, PE-PS and PP-*g*-PS were mixed together with PP/PS blends to study the efficiency of compatibilization in terms of dispersion and decrease in particle size of the dispersed PS phase. SEM showed the most advantageous outcome to the PP-*g*-PS and PE/PS copolymer with the best results coming from PP-*g*-PS.

The morphology of a PP/PE blend and maleic anhydride modified PP and PE (PP*g*/PE*g*) blend was studied by A. Colbeaux et al. (76). The initial morphology, at the extruder die, after the melt blending into a twin-screw extruder, was first characterized. The evolution of this initial morphology was followed after an injection molding operation, and during annealing at 200°C. The influence of the compatibilization of the polymers by coupling reactions through covalent and ionic reactions was also investigated. The ionic coupling additive showed good stabilization of the morphology. The coupling reactions delayed the occurrence of the coalescence and limited its effects. The extent of the coarsening depended strongly on the blend composition and the nature of the coupling agent. Still, the ionic agents appeared more effective.

The effects of PP-*g*-MA as compatibilizer on rheology, thermal, mechanical properties and the morphology of PP/PET blends were studied by H. Ebadi et al. (77). The SEM micrographs showed that the interfacial interactions were significantly improved by the addition of small amounts of the compatibilizer. A sharp decrease in the size of minor phase inclusions was observed, and the interface was saturated with compatibilizer and there was a significant improvement of the properties of the blends.

Z. Yu and M. Kancheng (78) established that reactive compatibilization with copolymers of the PP/PS blend improves the uniformity of the phase morphology, and strengthens the interfacial adhesion between the phases and the physical and mechanical properties.

Y. H. Na et al. (79) showed that the obtained data from the investigation of the morphology dependency on composition in the case of polylactide PLA-polycaprolactone PCL blends (compatible only at some extent), is a component ratio to cause the synergism of compatibilizing before apparent phase separation and coarsening. From the results of heat treatment, it is suggested that the control of domain sizes, as well as a PCL crystal ordering, can be achieved by coupling the phase separation with crystallization steps in PLA/PCL binary blends.

Z. Ailing et al. (80) revealed that a liquid crystal ionomer (LCI) affects, to a great extent, the miscibility, crystallization, and mechanical properties of PBT/PP blends. Increasing the LCI amount improves the miscibility of these polymers and only 1% of the compatibilizer increases some of the mechanical properties.

G. Bayram et al. (81) produced SMA copolymer-PE blends in a Brabender batch mixer and in a twin-screw extruder. As a compatibilizer, a PE with reactive epoxy functionality, and a nonreactive PE were used. At 25 and 50%, reactive PE contents the blends are considered compatible whereas, those with nonreactive PE were incompatible.

The blends were characterized in terms of morphology, viscoelastic and mechanical properties. The reactive blends have finer morphology than the nonreactive ones at all concentrations of the modified PE. The reactive blends also have higher modulus of elasticity, tensile strength, and strain at break than the nonreactive blends. The difference between the stress-strain behavior of the reactive and nonreactive system was most significant at 25% modified PE content.

Reactive compatibilization by the formation of *in situ* grafting of PBT with EVA-g-MA takes place competitively and simultaneously with the crosslinking of EVA and grafting of MA and EVA. It was considered that the maximum formation of the PBT-g-MA-EVA takes place at a certain MA content, producing the best reactive compatibilization in the PBT-EVA-g-MA blend and thus, the highest mechanical properties (82).

G. Radonjic and N. Gubeljar (83) investigated the compatibilizing effect of block copolymers such as SBS, SEBS, and SEP on the morphology and mechanical properties of immiscible blends produced from recycled LDPE and HDPE with 20% PS using SEM. It was found that the used copolymers were efficient in reducing the size of the dispersed phase and improving adhesion between PE and PS phases. SEBS was the most efficient compatibilizer.

M. Abdouss et al. (84) studied the effects of compatibilization of oxidized PP on blends made of PP/PA6 and PP/talc.

Phthalic anhydride terminated PS (PS-An) and S-MA copolymers were compared as compatibilizers and effect on morphology at low loadings (<10 wt%) in 70/30 PA66/PS blends. From the comparison of the PS particle sizes and the estimation of interfacial stability by observing (SEM) micelles, it was concluded that the higher MW PS-An is the most effective compatibilizer, showing finer morphology and higher interfacial stability (26).

The effect of the sulfonate group and counterion type on compatibility was evaluated by blending sulfonated and nonsulfonated forms of the amorphous polyester ionomer with both PA66 and PET. The melting point and phase behavior of the blends were determined by DSC and environmental SEM. A comparison between melting behavior between the melt and solution blends suggests that the compatibility is due to the specific interactions for ionomer/PA66 blends and transesterification for the ionomer/PET blends. The phase morphology of the melt blends was consistent with the results obtained by DSC analysis (85).

The miscibility, morphology, and tensile properties of a ternary blend system of PCL with PVC and with two chlorinated CPVC with different chlorine amounts (63 wt% and 67 wt%) have been studied by F-C. Chiu and K. Min (86). It was concluded that the interaction strength between CPVC and PCL follows the following sequence: CPVC67/PCL > CPVC63/PCL > PVC/PCL. The morphology of these blends showed little difference. They all had single-phase morphology in the amorphous regions and the crystalline blends exhibited birefringent spherulitic patterns.

The morphology and mechanical properties of PVC/SMA-g-PA6 blends were investigated by L. Dong et al. (87). SMA-g-PA6 was prepared via solution graft reaction between SMA and PA6. DSC analysis shows that the graft copolymer has a lower melting point of 187°C, which may result in a decrease in crystallinity of PA6 and thus, enable efficient blending of the grafted copolymer with PVC. Compatibilization was evidenced by the significant increase in mechanical properties, smaller particle size and finer dispersion of PA6 in PVC matrix, and, further, a continuous morphology at 16 wt% SMA-g-PA6 content. The authors found also that SMA-g-PA6 from the solution graft reaction is able to toughen and reinforce PVC.

W. Thielemans and R. P. Wood (88) found that butyrate kraft lignin improved the interface between an unsaturated thermosetting resin and reinforcing flax fibers. SEM images illustrate improvement in the adhesion of the resin to the fibers by showing the fibers fracturing together with the resin without fiber pullout.

The spontaneous or forced generation of particle-in-particle phase morphology during reactive processing of PBT with ethylene-methyl acrylate-glycidyl methacrylate (E-MA-GMA) random terpolymer has been investigated using different blending sequences (89). The obtained data demonstrated that the dimensions of this complex microstructure is not only controlled by the rate of interfacial grafting but rather by the relative kinetics between:

- the coalescence of the dispersed particles, and
- the interfacial compatibilization and/or elastomeric phase crosslinking.

Melt mixed poly(3-octylthiophene), POT and PVC are immiscible, but a co-continuous morphology is obtained with a relatively low content of POT. It is suggested that such a morphology is due to a low interfacial tension together with the difference in viscosity between these two polymers. The adding of DOP, a plasticizer for both polymers is able to further decrease the interfacial tension, and a co-continuous phase is obtained (90).

In order to examine the influence of strong intermolecular hydrogen bonding on the dynamic of polymer blends, S. H. Zhang et al. (91), using broadband dielectric relaxation spectroscopy (DRS), studied miscible poly(4-vinylphenol)/poly(vinyl methyl ether) PVPh/PVME mixtures with PVPh concentrations up to 80%. Different relaxations behaviors were observed for blends with different compositions, and thermorheological simplicity does not exist in this dynamically asymmetric blend, despite the presence of strong intermolecular interactions.

Blends of thermoplastic PU (TPU) and EPDM were prepared via melt blending, and morphology, mechanical properties, and rheology were studied by W. Xiaodong and L. Xin (92). SEM micrographs demonstrated that a network of EPDM was formed in TPU matrix, and became finer and more perfect with the addition of 8 wt% EPDM. DMA and FTIR spectroscopy indicated that EPDM was thermodynamically miscible with the soft segments of TPU and incompatible with the hard segments. Rheological properties revealed that the introduction of EPDM into TPU resulted in a reduction of viscosity at a high shear rate and a decrease of the flow activation energy; thus, the processability of the blend was improved.

The morphology of immiscible and highly incompatible blends of poly(trimethylene terephthalate)-PTT and EPDM blends was studied by I. Aravind et al. (93), with and without the addition of the compatibilizer maleic anhydride grafted ethylene-propylene elastomer (EPM-g-MA). The addition of this compatibilizer to the blends tends to decrease the free volume showing its compatibilizing effect.

The morphologies of NBR/EPDM blends by the combination with mercapto and oxazoline groups were examined through SEM. Blends compatibilized with the EVASH-based systems showed finer morphologies than the non-compatibilized blend of those compatibilized with EPDMSH-based systems. In non-vulcanized NBR/EPDM (70/30 wt%) blends, the presence of the co-reactive EVASH/NBR-ox system changes the morphology from a continuous structure towards a droplet-matrix morphology, and also results in a significant broadening of the damping curve related to NBR transition, observed from DMAT analysis. The effect of the compatibilization on aging has also been evaluated (45).

Conclusions

Polymer blending is one of the most effective and economical techniques used to obtain, with a lower cost, new materials with distinct properties than those of each blend component. This technique is cheaper than the synthesis of new monomers and/or new polymerization ways in view to produce new polymeric materials.

It is known that most polymers are thermodynamically immiscible, and the blend components usually phase-separate into distinct macroscopic domains, and consequently, show poor properties. The introduction in the blend system of compatibilizers makes it possible to produce polymeric materials with desirable properties.

Compatibilization of polymer blends continues to be a fruitful strategy of research in the polymer industry. This research is focusing on applications of compatibilizer technology such as the recycling of post-consumer plastics without extensive sorting, the production of multi-layer film, packaging products, as well as the development of new materials based on plastic/plastic, elastomer/elastomer, and plastic/elastomer blends.

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