CHAPTER – 1

INTRODUCTION
TO
POLYMER BLENDS

CONTENTS

Plastics have enjoyed extensive growth during past two decades. They have replaced metals, glass, ceramics, wood and natural fibers in wide variety of industries. Success of various polymers in such a broad spectrum of applications is attributed to a wide range of properties available with polymeric materials[1,2]. Earlier the development in polymeric materials involved synthesis of new homopolymers and copolymers for tailoring the properties. Now, realisation is that,

- new molecules are not always required to attain desired properties,
- secondly newer polymers with desired properties are not synthesized on a routine basis and
- ***blending/alloying*** can offer the opportunity to develop materials that might even show synergism in properties[3].

For engineering applications, it is generally desirable to develop easily processible polymers that are dimensionally stable, can be used at high temperature and also resist attack by solvents or by the environment. ***Blending/Alloying*** offers ease of processing and the method is cost effective, hence has enormous growth potential for speciality engineering applications[4].

Basically, polymer blends are nothing but it is a mixtures of chemically different polymers and/or copolymers. They are mainly systems with the multiphase structure, while alloys are a class of polyblends in which a large interpenetration of domains is secured by either chemical or physical means. Blends are direct result of the blending action, while alloys are the final blends of well defined morphology and set of properties. The primary advantages in the use of polymer blends and/or alloys are

a) better performance at a reduced price
b) expansion of the performance of speciality resins
c) reuse of plastic
d) generation of unique materials with respect to process ability and/or performance

Polymer blends can be either miscible or immiscible. The miscible blends are further divided into two sub classes.

(i) Homogeneous blends

It is a mixture of chemically identical polymers differing in molecular masses e.g., blend of low and high molecular weight polystyrene(PS)

(ii) Heterogeneous blends

It is a mixture of two or more than two chemically different polymers e.g., blend of polyphenylene ether (PPE) and polystyrene

Miscible systems exhibit a single glass transition temperature (Tg) located between the Tg values of the two components and is expected to behave like single component. They are generally transparent and can be moulded without streaking. The heat distortion temperature (HDT) of miscible blends varies smoothly with composition and thus is easy to predict and control[5,6]. While in an immiscible polymer blend which forms multi-phase system there exists composition gradients whose level depends on the intensity of mixing and on solubility parameter values (δ) of the polymers. Hence a third component is required (i.e., compatibilizer or interfacial agent) which can reduce the degree of 'immiscibility' or improve the 'compatibility' by mechanical or physical means. It also helps in decreasing the interfacial tension and provides the morphological stability[2]. Compatibilizers are generally block, graft or ionomeric copolymers of mutually compatible polymers and lead to formation of alloys where interpenetration of domain is achieved to great extent. Fig.1.1 illustrates a broad classification of polymer blends and /or alloys. Utracki[6] has suggested a few guidelines for designing a polymer blend which is illustrated schematically in Fig.1.2.

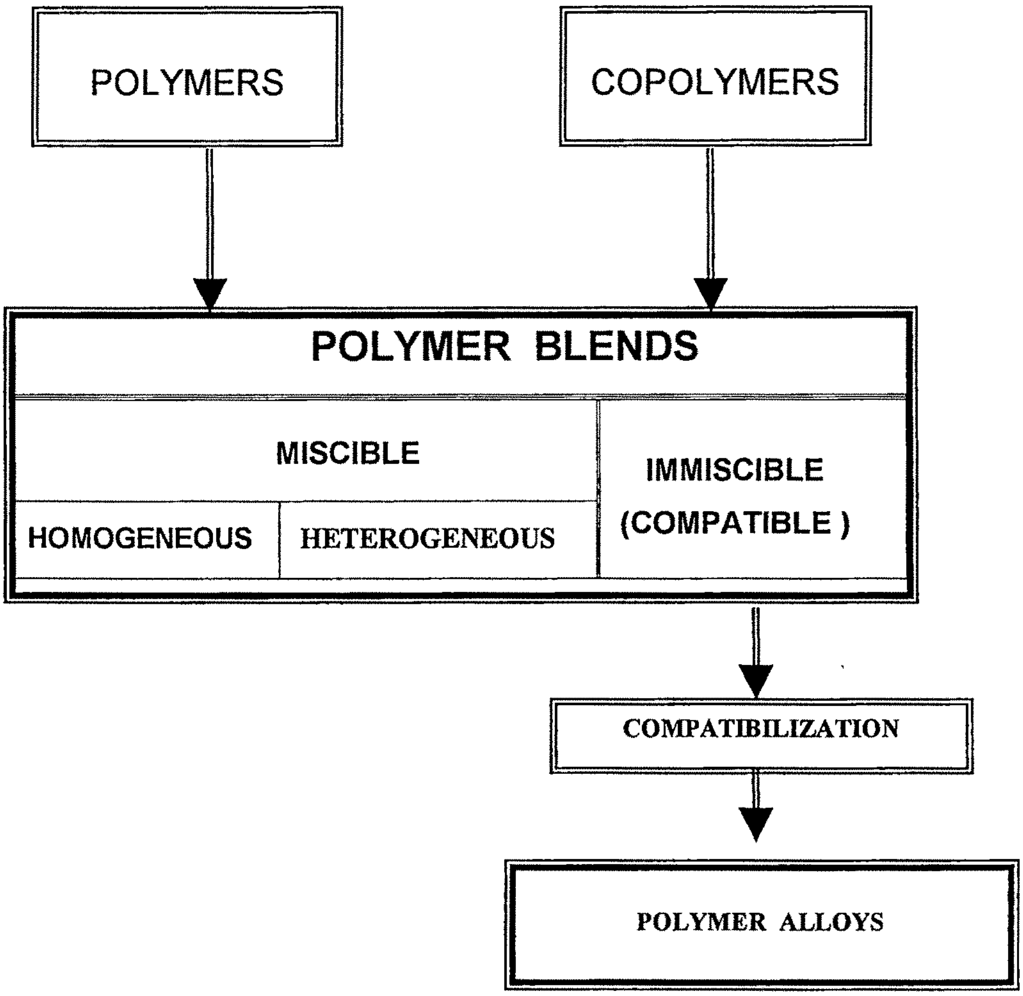


Figure 1.1 : Interrelations in polymer blend nomenclature

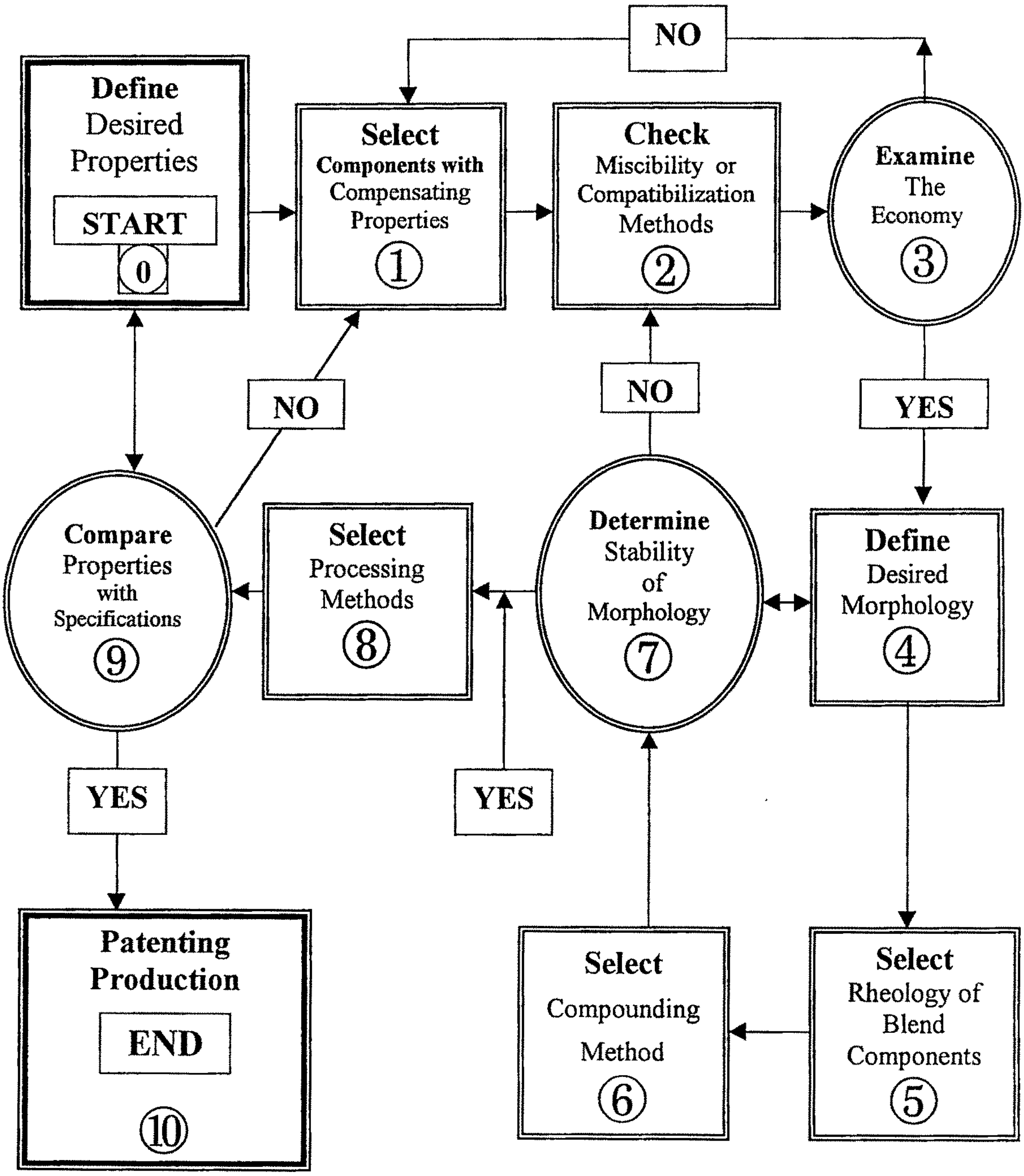


Figure 1.2 : **Schematic representation of the steps to be taken while developing polymer blends and alloys**

1.1.1 Polymer blend terminology[7]

- ***Polymer :*** A substance composed of large molecules, the macromolecules, built by co-valently joining at least 50 molecular mers, segments or recurring constitutional repeating units. Commercial polymers may contain upto 2 wt % of another polymeric modifier.
- ***Copolymer :*** Polymeric material synthesized from more than one monomer.
- ***Engineering polymer :*** Processable polymeric material, capable of being formed to precise and stable dimensions, exhibiting high performance at the continuous use temperature above 100 °C, and having tensile strength in excess of 40 MPa.
- ***Polymer blend :*** Mixture of at least two macromolecular substances, polymers or copolymers, in which the ingredient content is higher than 2 wt %.
- ***Homogeneous polymer blend :*** Mixture of two homogeneous polymers, usually with narrow molecular weight distribution fractions of the same polymer.
- ***Miscible polymer blend :*** Polymer blend, homogeneous down to the molecular level, in which the domain size is comparable to the macromolecular dimension, associated with a negative value of the free energy and heat of mixing, $\Delta G_m = \Delta H_m < 0$, and $d^2\Delta G_m/df^2 > 0$. Operationally, it is a blend whose domain size is comparable to the dimension of the macromolecular statistical segment.
- ***Immiscible polymer blend :*** Polymer blend whose free energy of mixing $\Delta G_m = \Delta H_m > 0$.
- ***Compatible polymer blend :*** Term to be avoided ! At best a utilitarian, non-specific term indicating a marketable, visibly homogeneous polymer mixture, with enhanced performance over the constituent polymer.
- ***Polymer Alloy :*** Immiscible, compatibilized polymer blend with modified interface and morphology.

- ***Compatibilization :*** Process of modification of the interfacial properties in an immiscible polymer blend, resulting in formation of the interphase and stabilization of the desired morphology, thus leading to the creation of a polymer alloy.
- ***Interphase :*** Third phase in binary polymer alloys, engendered by inter diffusion or compatibilization. Thickness of the layer varies from 2 to 60 nm, with the type of polymers and compatibilization method.
- ***Compatibilizer :*** Polymer or copolymer that added to a polymer blend which modifies its interfacial character and stabilizes the morphology.
- ***Chemical compatibilization :*** Compatibilization by addition of a compatibilizer, in most cases either a copolymer or multi polymer.
- ***Physical compatibilization :*** Compatibilization by physical means e.g., high stress field, thermal treatment, irradiation, etc.
- ***Reactive compatibilization :*** Compatibilization during reactive processing, extrusion or injection molding.
- ***Engineering polymer blend :*** Polymer blend or polymer alloy either containing or having properties of an engineering polymer.
- ***Interpenetrating polymer network (IPN) :*** Polymer alloy, containing two or more polymers in network form, each chemically crosslinked. Sequential, simultaneous (SIN) and latex type IPNs are also known.
- ***Thermoplastic IPN :*** Polymer alloy, containing two or more polymers physically crosslinked in a co-continuous network form. The crosslinking originates in crystallinity ion cluster formation, presence of hard blocks in copolymers, etc.

1.1.2 Methods of blending [8]

1.1.2.a Solution blending

Two polymers may be thermodynamically miscible but physically incompatible due to large difference in the melt viscosities at conditions of compounding. In such cases, solution blending is the alternate choice.

Secondly, it is also useful if polymers show degradation under melt processing conditions. In solution blending both polymers are dissolved in the same or miscible solvents separately and then mixed thoroughly. However, removal of the diluent can often lead to changes in the domain size of the blend components leading to polymer separation. As solvent is removed from a one-phase mixed polymer, solution contacts of dissimilar solvated polymers will bring about phase separation. Though It is not an economically viable process it is useful for preparing films/membranes (e.g., polysulfone blend with polyvinylpyrrolidone).

1.1.2.b Latex blending

Latex mixing is one of the most important techniques for the preparation of commercial polyblends. In latex blending, polymers are present as suspended micro-phase. Interactions of neighboring spheres are prevented by the suspending medium. The disadvantage of this technique is the increase of cost and environmental hazard e.g., polymerization of vinyl chloride in the presence of polybutyl acrylate, rubber modified epoxy resins, ABS etc.

1.1.2.c Melt blending

This technique involves mixing of two polymers in the molten state under shear. "*Brabender Plasticoder*" type batch mixer or an extruder (single or twin screw) is used for blending. Melt blending is used for a system in which thermal degradation does not occur at the processing temperature. Melt blending has several advantages over solution as well as latex blending. In melt blending, liquid or dispersion agent is eliminated, which reduces costs associated with solvent removal, recovery and losses. Additionaly, combination of only the elements desired in the final mixture, reduces the likelihood of contamination in the alloys or blend. Although, this technique has some disadvantages like, degradation of either or both polymers can occur, and crosslinking, block, graft formation or chain scission reactions can result. Because of these degradative conditions blended polymers are often physically different from what might be expected for the composite. This can be avoided by using heat sensitive polymeric plasticizer of low molecular weight whose temperature is well below the degradation point of either polymer, which suffices to give a mixture at low enough viscosity so that primary bond cleavage from shear forces is avoided. Most of the blends have been prepared by melt mixing technique. Widely used PP/Polyamide, PP/PC, PP/ABS, PP/AE, ABS/PVC blends are prepared by melt mixing technique.

1.2 Theoretical background of factors affecting the polymer blend properties

1.2.1 Phase behaviour in polymer blend

The phase diagram for a strictly two component system, i.e., a mixture of two monodispersed polymers, could resemble any of those in Fig.1.3, in which systems with either an upper critical solution temperature (UCST) or a lower critical solution temperature (LCST) or both or with a tendency toward both are shown. Even though synthetic polymers are never monodispersed, they exhibit phase diagrams that resemble these with some variations, with the exception of the system in Fig.1.3c. In addition, two-peaked coexistence curves have been observed in the presence of solvents for certain polymer mixtures such as polystyrene-polybutadiene[9] and for the mixture of styrene and isoprene oligomer in the absence of solvent[10] (Fig.1.4). Mixtures that have positive (endothermic) heats and entropies of mixing usually tend to exhibit UCST where as mixtures that have negative (exothermic) heats and entropies of mixing usually exhibit LCST.

Fig.1.3 and 1.4 can be used to demonstrate a dilemma that affects any discussion of miscibility or compatibility. At temperature above UCST or below LCST, the binary mixtures are completely miscible at all compositions.

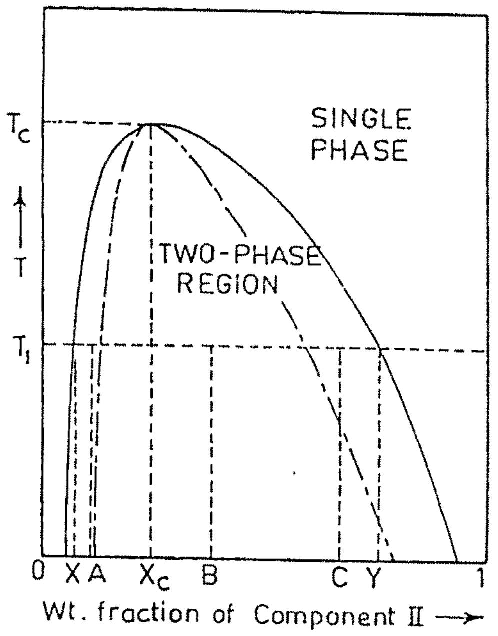


Figure 1.3a

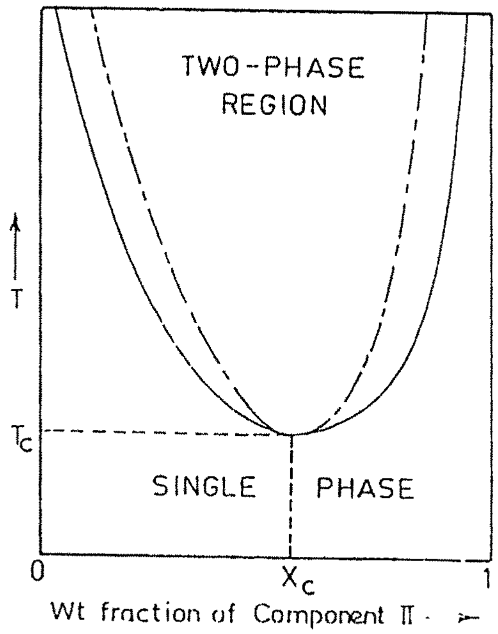


Figure 1.3b

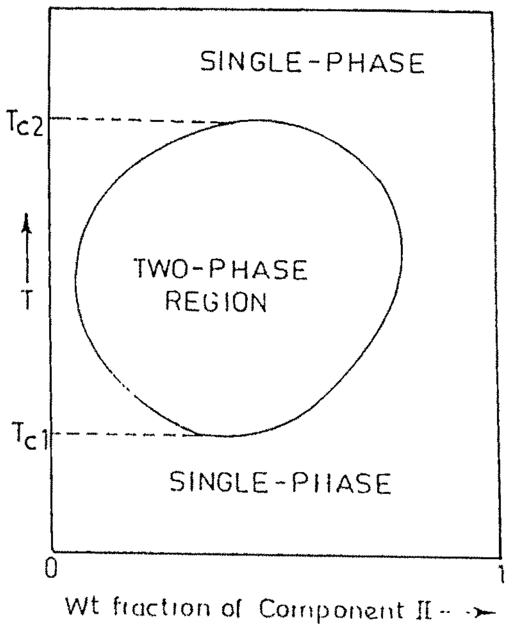


Figure 1.3c

Figure 1.3 : Phase diagrams for various mixtures as a function of temperature

a) mixture with an UCST : (——) binodal; (– – –) spinodal

b) mixture with a LCST : (——) binodal; (– – –) spinodal

c) A mixture with an UCST above a LCST

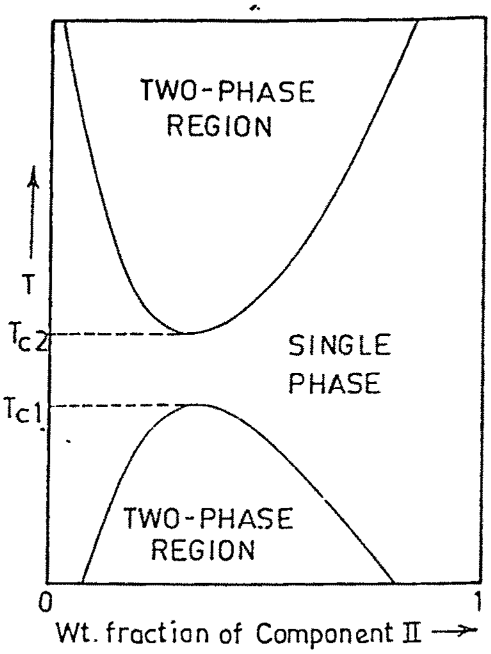


Figure 1.3d

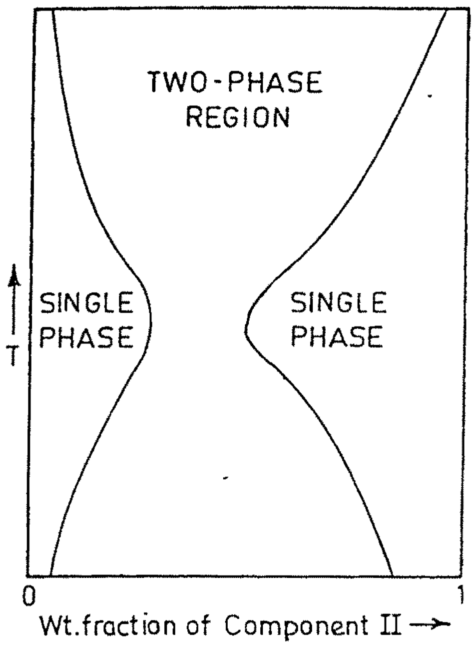


Figure 1.3e

d) A mixture with a LCST above an UCST

e) A mixture with a tendency toward greater solubility of intermediate temperatures

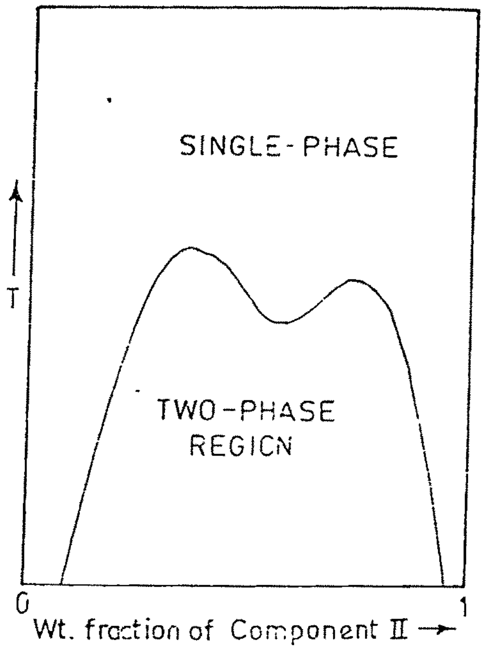


Figure 1.4 **Two-peaked coexistence curve as seen in the case of polystyrene-polybutadine-solvent mixtures and for mixtures of styrene and isoprene oligomers.**

Below UCST and above LCST, there are still compositions, generally those in which one of the components is present in small percentages in which only a single phase is observed, while at intermediate compositions phase separation occurs, and such mixtures are very important. In such mixtures the boundary between stable and metastable compositions is called binodal and the boundary between metastable and unstable compositions is called spinodal. Compositions along the binodal are the ones that define the phases into which unstable and metastable compositions will separate, e.g., in Fig.1.3a, at temperature T_1, compositions A, B, and C will separate into compositions X and Y. The mixture forms a single phase whenever its composition is outside the range X-Y. Tc and Xc are critical temperature and composition, respectively. For a binary system, a critical point indicates the circumstances under which the system will just begin to separate into two phases, which can be called as limit of compatibility. If two polymers are incompatible in the molten phase then it is not possible to produce a homogeneous blend by melt mixing technique. However, it is possible to produce a solution blend at a temperature below LCST. In such cases where the LCST curves lie at a temperature below the melting point, it is important to ensure that the blend is not used in the application, where the material would be exposed to temperature above the LCST curves[11].

1.2.2 Thermodynamics of blending

For the discussion of thermodynamics of polymer mixture, one can take the Flory-Huggins free energy of mixing as the starting point. The compatibility of the blend components can be expressed thermodynamically by change in free energy, during mixing of polymeric components and is expressed as

$$\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix} \qquad (1)$$

where, ΔG_{mix} = free energy of mixing, ΔH_{mix} = enthalpy of mixing,
ΔS_{mix} = entropy of mixing and T = temperature

Thermodynamically the mixing process will only be favored if ΔG_{mix} is negative.

When two different polymeric molecules have little attraction for each other, the enthalpy of mixing (ΔH_{mix}) is positive and it is an unfavorable condition for compatibility between two phases. Secondly when these polymeric molecules are mixed, the mixture is less random due to the restricted movement of atoms of polymeric chain resulting into lower value of entropy of mixing (ΔS_{mix}). As a result the value of - $T\Delta S$ will be insufficient to overcome the positive enthalpy of mixing (ΔH_{mix}), which results in the positive value of ΔG_{mix}. As a result the two polymers become incompatible with each other.

Several quantitative theories have been proposed to estimate ΔG_{mix}, ΔH_{mix} and ΔS_{mix} for polymeric mixture[12-14]. However, there is no sufficient experimental data to verify which of these theories are adequate[3,15]. Flory-Huggins-Staverman theory of thermodynamics of polymer mixture, which is based on lattice model[16] and statistical thermodynamics[17], has been observed to be more suitable in describing many of the qualitative features of polymer blend thermodynamics. The FHS expression for the free energy of mixing of two polymers is :

$$\Delta G_{mix}/ VRT = \phi_1 / v_1N_1 \ln \phi_1 + \phi_2/ v_2N_2 \ln \phi_2 + \phi_1\phi_2 \chi / \nu \qquad (2)$$

where, V = total volume of sample

R = the gas constant

T = the absolute temperature

N_i = the degree of polymerization of ith component.

ϕ_i = the volume fraction of ith component

v_i = the molar volume of ith mers

ν = an arbitrary volume

In above equation the first two terms represent the combinatorial entropy of mixing, and the last term comes from the interaction enthalpy χ called as *"Flory interaction parameter"*.

Most polymers of commercial interest have degree of polymerization of 1000 or more and the entropy of mixing is quite small. This low combinatorial entropy of mixing can be thought of as a direct result of the high configurational entropy that is characteristics of polymer chains[18]. The entropy of mixing depends on the mole fraction of chains, rather than the volume or weight fraction and hence the first two terms in Eq-2. are divided by the degree of polymerization of the two components. The interactions between the two polymers are quite similar to those between their small molecules. The forces of interaction are quite local, extending only over a range of the order of a repeating unit, so the enthalpy of mixing will not depend to any significant amount on the molecular weight of components. But for most mixtures, the enthalpy of mixing is positive. The entropy of mixing of small molecules can be so large that it over comes completely the positive mixing enthalpy and so causes the components to mix under a wide range of temperature and composition. As according to the above discussion regarding the changes in entropy as well as enthalpy values during mixing of two polymeric components they could not show an essential region of miscibility.

In the prediction of blend miscibility through FHS-theory it is needed to determine the conditions under which a blend can become miscible. There are basically three general classes of miscible blends based on the manner in which miscibility is achieved.

(i) In polymer blends consisting quite low molecular weight component the entropy of mixing is large enough to out balance the enthalpy of mixing. From FHS theory, one can derive the following expression for the critical composition and temperature of the blends:

$$\phi_{1crit} = 1 / [1 + (N_2v_2 / N_1v_1)^{1/2}] \qquad (3)$$

$$\chi_{crit} = V / [2 (1/N_1v_1 + 1/ N_2v_2)^2] \qquad (4)$$

Blend is symmetric if both components have similar parameters, i.e., if $N_1=N_2=N$ and $v_1=v_2=v$. In that case $\phi_{1crit} = 0.5$ and $(\chi_N)_{crit} = 2$. In most cases the components of a polymer blend are not very different from one another in molecular weight or density. Therefore, in general these relations for the symmetrical blends are good rules of thumbs for blends. So even if χ is large for a particular blend, if **N** is small enough, the blend can be miscible. But, if the molecular weight is very low, then many of the desirable properties of the blends will be lost, hence the use of low molecular weight polymer is not an effective strategy for achieving miscibility in blend. Only when the interactions are very weak as in the case of dispersive Vander Waals force then this might be worthwhile[19].

(ii) The other type of blends have negative value of χ and so the enthalpy. This is due to the presence of strong attractive interactions between the blend component, such as hydrogen-bonding[20] between donating and accepting groups, ionomeric interactions[21,22] and acid-base interaction[23] which can also induce the mixing of the polymers.

(iii) Third type of blends have negative value of χ, but not because of any special attractive interaction between the components (i.e. intra or inter molecular), but one or both of the components are statistical copolymers[24] and the balance of the forces among the several monomer units results in miscibility. This has been termed as "copolymer effect."

Although FHS-theory explains many of the general observations of immiscibility in blends. There are following deficiencies in it.

(i) It can not quantitatively describe blends with a significant degree of miscibility.

(ii) It can not describe the behavior of blend with an LCST phase diagram.

(iii) The theory predicts that χ only depends on temperature and is independent of molecular weight or composition. But recent studies have shown that there is a clear compositional dependence of χ, even in blends when only dispersive forces are operating and in blends of a polymer with its isotopic twin.

1.2.3 Interfaces in polymer blends

It is almost universally accepted that blending produces crumbly "*cheesy*" materials with no useful mechanical properties. This behaviour stems from the immiscibility of the polymers, with the association of like molecules and repulsion of dissimilar molecules. The net result is a lack of adhesion between phases.

Unfavorable enthalpies of mixing tend to minimize contacts between unlike polymers and the interpenetration of dissimilar molecules at the polymer-polymer interface. Minimum contacts correspond to an absolutely sharp interface with no interpenetration of molecules. The unfavorable enthalpic interactions which drive the immiscibility mean that there will be repulsive interactions at the inevitable interface which will counteract the weak Vander Waal's interaction and help to create the lack of adhesion. In combination of hydrophilic and hydrophobic materials poor adhesion may arise because of unfavorable interactions[25].

In polymer blends the polymer-polymer interface is not totally impenetrable. Any exploration of chains across the interface will reduce the entropy loss but introduce enthalpic interactions. A balance of these two will be established with some interpenetration and the development of

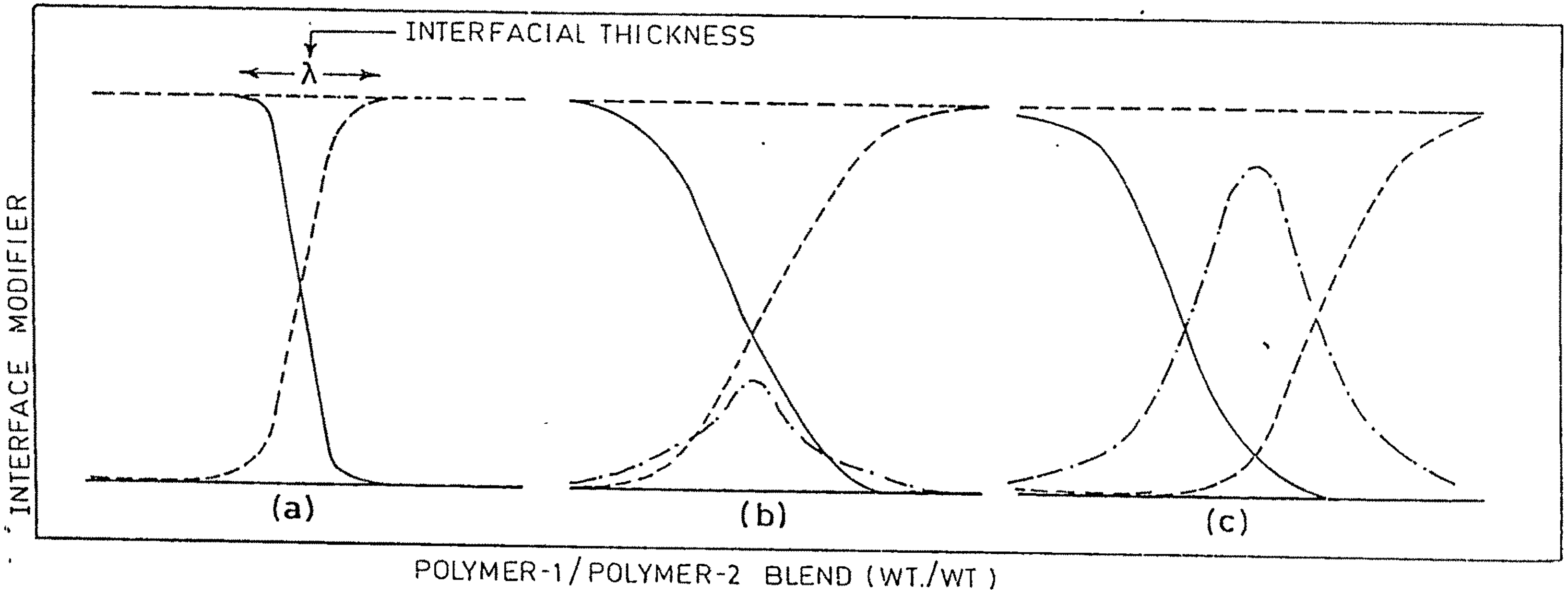


Figure 1.5 : Schematic representation of concentration profiles in a polymer-1/ polymer-2 blend

a) a sharp interface (polymer-1 : ——, polymer-2 : - - - - -)

b) an interface modified with a low molecular weight copolymer (–·–·–·–·–)

c) an interface modified with a high molecular weight copolymer (—·—·—·—)

concentration profiles across the interface, as illustrated in Fig.1.5. Theoretical calculations of these profiles have also bean performed[26].

So to enhance interfacial adhesion it is necessary to modify the interface and create links across the interface. For this chains located at the interface require to interact favorably which can be achieved by the incorporation of an interfacial agent, which may be either block copolymer, graft copolymer or ionomer. Thermodynamically it can be considered to change enthalpy or entropy as discussed in 1.2.2.

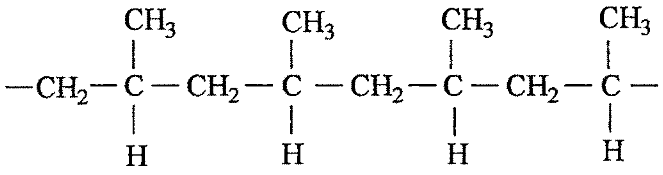
1.2.4 Blend morphology

The properties of blends/alloys depend very significantly on the morphology produced during processing[27]. The tailoring of blends to achieve desired properties requires an excellent control of morphology of the phases and their stability once formed. The arrangement may consist of one phase dispersed as simple spheres in a matrix of the other polymer. The major component becomes the continuos phase and the size and shape of the droplets of the dispersed minor phase depend upon the interfacial tension between the phases, viscosity, elasticity and the deformation field imposed. The shape of the dispersed phase can change from spheres to fibrils by uniaxial elongation flow as in fiber spinning or into platelets by biaxial stretching as in blow moulding. The dimension of the phases are also important in all these morphologies. Another distinct morphology consists of both phases simultaneously having a continuos character or an interpenetrating network of phase. The morphology of incompatible blends can be altered significantly by the addition of compatibilizers. Compatibilizer can significantly reduce the size of the domains in the blend and make the morphology less sensitive to processing. The compatibilizers reduce the interfacial tension and improve adhesion between the phases producing an interpenetrating polymer matrix in some cases.

1.3 Polypropylene (PP) and acrylonitrile-butadiene-styrene (ABS)

Polypropylene

The monomer is produced by the cracking of petroleum products such as natural gas or light oils. For the preparation of PP the C_3 fraction (i.e., propylene and propane) is the basic intermediate, which is separated from the other gases by fractional distillation. A typical catalyst system may be prepared by reacting titanium trichloride ($TiCl_3$) with aluminum triethyl, aluminum tributyl or aluminum diethyl mono chloride in naphtha under nitrogen to form a slurry consisting of about 10% catalyst and 90% naphtha. The properties of the polymer are strongly dependent on the catalyst composition and its shape and size. Commercially it is developed by suspension process. Atactic, syndiotactic and isotactic forms produced during processing are shown here.



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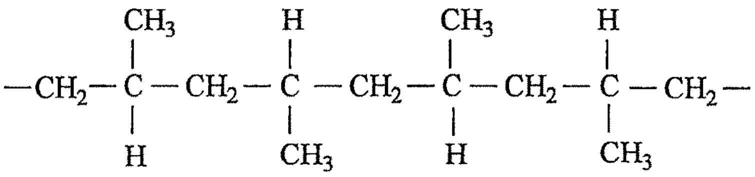
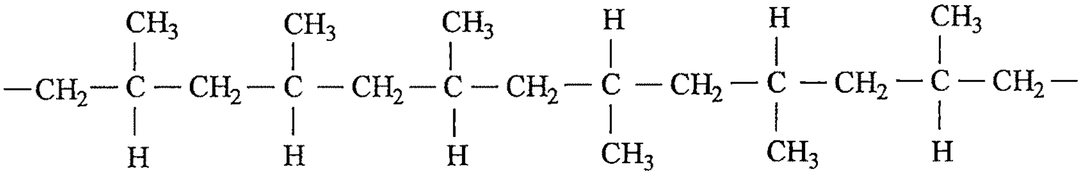
Isotactic polypropylene



$$-CH_2-\underset{\displaystyle H}{\overset{\displaystyle CH_3}{\underset{|}{\overset{|}{C}}}}-CH_2-\underset{\displaystyle CH_3}{\overset{\displaystyle H}{\underset{|}{\overset{|}{C}}}}-CH_2-\underset{\displaystyle H}{\overset{\displaystyle CH_3}{\underset{|}{\overset{|}{C}}}}-CH_2-\underset{\displaystyle CH_3}{\overset{\displaystyle H}{\underset{|}{\overset{|}{C}}}}-CH_2-$$

Syndiotactic polypropylene



$$\begin{array}{ccccccccccccc} & CH_3 & & CH_3 & & CH_3 & & H & & H & & CH_3 & \\ & | & & | & & | & & | & & | & & | & \\ -CH_2- & C & -CH_2- & C & -CH_2- & C & -CH_2- & C & -CH_2- & C & -CH_2- & C & -CH_2- \\ & | & & | & & | & & | & & | & & | & \\ & H & & H & & H & & CH_3 & & CH_3 & & H & \end{array}$$

Atactic polypropylene

Polypropylene is a crystalline liner polymer, containing little or no unsaturation. Propertywise, it has higher softening point and hence provides higher performance temperature, good stability on environmental stress cracking problems and good chemical resistance. However, it is susceptible to oxidation and toughness with low cost. It can be used for film, fiber and moulding process[28].

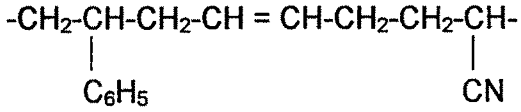
Acrylonitrile-butadiene-styrene (ABS)

ABS is a very versatile family of engineering thermoplastics produced by combining three monomers, acrylonitrile, butadiene, and styrene. A very few engineering thermoplastic families can offer a line of products meeting the needs of the vast number of applications for which ABS has been found suitable. ABS plastics offer a unique combination of toughness, rigidity and quality surface appearance along with the ability of process for thermoplastic processing. It is also useful for load bearing application due to its excellent dimensional stability and low creep properties. This broad range of ABS applications is due to the wide variety of products, which can be produced from the uniquely versatile ABS morphology. ABS consists of two phases, a continuos phase of styrene-acrylonitrile (SAN) copolymer and dispersed phase of grafted polybutadiene particles. Each of these phases contribute unique characteristic to the polymer.

Each of the monomers, acrylonitrile, butadiene and styrene is an important component of ABS and each monomer is having its own speciality.

Styrene balances clarity, rigidity, ease of processing and relatively low cost. Acrylonitrile has very good solvent resistance, better surface hardness and improved toughness. Butadiene provides flexibility, which results into toughness and good impact resistance.

Structure of ABS :



$$-CH_2-\underset{\underset{C_6H_5}{|}}{CH}-CH_2-CH=CH-CH_2-CH_2-\underset{\underset{CN}{|}}{CH}-$$

In the process, the grafted polybutadiene phase and the continuous SAN phase are both polymerized separately and then melt compounded along with stabilizers, lubricants, and colorants. Emulsion polymerization technique is commonly used to produce the grafted polybutadiene phase. This grafted polybutadiene emulsion latex can be coagulated to form ABS as a powder, which contains a higher level of rubber. This ABS-powder can further melt compounded with SAN to form finished ABS products, which contains various levels of rubber portion[29].

1.4 Literature survey on polypropylene blends with engineering plastics

1.4.1 PP/ABS blends

Studies on the blends of polypropylene (PP) with acrylonitrile-butadiene-styrene tercopolymer (ABS) are mostly confined to patents and rarely to publish literature. Kamosaki et al.[30,31] have studied the blending of ABS with crystalline polyolefins. Blends have been reported to have very good molten fluidity, rigidity, low shrinkage, and good coating and adhesion properties. In blending process, 70% PP, 30% ABS was blended at elevated temperature in extruder. Acrylonitrile and/or styrene grafted on rubber or acrylonitrile-styrene copolymer was used as a compatibilizer in small portions.

Melt index, flexural and impact strength were observed to increase, whereas % shrinkage was observed to decrease in blend when compared to pure polypropylene. They have also synthesized PP/ABS blends, using PP-g-MMA as a compatibilizer at 230°C. Mechanical properties of modified plastics were reported to improve[32]. Idemitsu Kasan. Co. Ltd.,[33] also studied the blends of PP/ABS/talc for improving the electrodeless plating properties of polypropylene. Markin and Williams[34] observed the better tensile strength of PP/ABS blends, when dispersion was better and better impact strength when dispersion was poor. Lamination of PP with ABS and/or vinyl polymer was studied by Ube. Industries Ltd.,[35] where ethylene-vinyl acetate copolymer, ethylene vinylacetate-vinylchloride, styrene-vinylacetate copolymer or chlorinated polypropylene was used as an adhesive. The modified plastic was used for furniture. Morphological and mechanical properties of PP/ABS/talc blend as an engineering plastic were studied by Wang and Zhu[36]. They have reported that modified material has good shaping properties, bending rigidity, deformation rebound elasticity and dimensional stability. Maleated PP/ABS base polymer blends were developed by Go et al.[37]. They have reported that the blends have high impact strength and heat resistance capacity. Gupta, et.al.[38] studied the impact and tensile properties of binary PP/ABS and ternary PP/ABS/LDPE blends. They have observed considerable impact toughening of PP on blending with ABS. The maximum impact strength was observed at 10 (wt %) of ABS in PP/ABS and PP/ABS/LDPE blends. They have also observed the shear-induced fracture with increased ductility with increasing ABS content in binary and ternary blends. The compatibility of blends of ABS resin and polyolefin especially PP was thoroughly studied by Ezaki and Aibe.[39]. According to their observation, polypropylene modified with maleic anhydride(MAH) and a polymer based on hydroxyl, cyano and/or aromatic group containing monomers became useful for improving the compatibility. In the process, reported by them, a mixture of 30% PP, 70% ABS, 7 parts, PP-g-MAH copolymer, 3 parts, acrylonitrile-2-hydroxypropyl methacrylate-styrene tercopolymer, and 1 part dimethyl aminoethyl methacrylate-styrene copolymer mixture was blended. The size of dispersed particles was reported to be 0.6 -

1.5 μm and izod impact values were observed to be 7.3 at 4 kg.cm/cm. ABS/PP bonded with acrylonitrile-glycidyl methacrylate-styrene copolymer as a compatibilizer with the weight % ratio of 23.1:1.5:75.5(ABS/compatibilizer/PP) was studied by Padwa[40]. He has observed that this composition will give a laminate with higher shear strength (4.6 MPa), while a blend with excellent impact resistance properties was developed by Aibe[41]. According to their process, 30% PP, 70% ABS, 40 parts, PP-g-MAH copolymer and 6 parts acrylonitrile-diethylaminoethyl methacrylate-styrene copolymer was kneaded palletized and injection moulded to less than 2.0 μm dispersed particle size. The chemical and heat resistance of ABS polymer could be enhanced by blending ABS with crystalline polyolefins as studied by Hidetoshi, et al.[42]. They used PP-base graft copolymer as a compatibilizer. The blends were reported to have very good solvent as well as heat resistance and good processability. Padwa,[43] has also studied the PP/ABS blend. According to his report ethylvinyl acetate (33.0 ± 5.0%) could work as an effective extrudable tie layer. The thermal, mechanical and morphological properties of PP/ABS blends were studied by Frounchi and Paul[44]. For achieving compatibility between these two styrenic copolymers and/or thermoplastic rubber base polymer compatibilizer was used. Blends for automobile applications particularly for bumpers, were developed by Oota[45]. The automotive interior trim thermoplastic resins based on PP/ABS were developed by Jones[46]. They studied the effect of shear rate and behavior on impact strength and stiffness of ABS and/or PP and concluded that shear rate and property showed no effects on stiffness of each material but it affects the impact strength.

Near IR spectroscopy (NIR), differential scanning calorimetry (DSC) and FTIR analysis as quantitative methods for compositional analysis of blends of recycled PP/ABS polymer were used by Walker and Sigbritt[47]. Morphological and mechanical properties of PP/ABS blends compatibilized with pp-g-acrylic acid were studied by Patel et al.[48]. They have observed that PP-rich ternary blends show good morphological and mechanical properties and the use of 5 wt % PP-g-acrylic acid as a compatibilizer resulted in fine and homogenous dispersion of ABS in the PP. It was observed that in PP/ABS (90/10) blend 2.5 phr PP-g-2-HEMA as a compatibilizer was sufficient for toughening effect. Impact strength was reported to be 1.2 kg cm/cm. At higher level of compatibilizer (7.5 phr) impact strength was reported to decrease. They have also examined the suitability of various existing theoretical models for the prediction of tensile moduli of the blends[49]. Vacuum-molded laminates of PP/ABS useful for automobile interior parts were developed by Araki and Tabata (Snigeru Col. Ltd.)[50]. According to their process a laminated sheet composed of a 0.4 - 1.6 mm thermoplastic polyoletin elastomer skin layer, a 1 - 4 mm PP foam as a cushning layer and 0.4 - 3 mm core layer containing 30 - 40% PP and 55 - 65% ABS showed good moldability. The effect of blend composition and compatibilizer on the mechanical properties of PP/SAN and PP/ABS blends was studied by Park et al.[51]. PP-g-styrene-acrylonitrile copolymer was used as a compatibilizer for PP/ABS blend, and 5 phr of compatibilizer could give maximum mechanical strength (flexural and tensile strength) and minimum droplet size.

1.4.2 PP/Nylon-6 blends

Polypropylene/polyamide blends belong to a large and growing family of polyolefins (PO) alloyed with polyamide (PA). Since 1985, PP has been used as PO with increasing frequency and nylon-6 as PA[52]. Other PO/PA blends, for example PP with PA - 6,6, PA - 12 or with aromatic polyamide are also available. The reasons for blending PO and PA are to bridge the property gap between the resins, to reduce the cost, to decrease the water absorption of PA, to improve processability of PP and to benefit from the thermal and mechanical properties of polyamide with improving its impact resistance. Laminated moulded plastic with improved layer bonding strength was prepared by blending a polypropylene with polyamide and/or polyester. PP-grafted with unsaturated carboxylic acid or its derivatives were used as a binder between PP/PA plastics[53,54]. Mitsui Toatsu Chemical Inc.,[55] has also reported the similar work and the laminated films developed were used for food packing. Hayashida and Yoshida[56] used an extruder type capillary rheometer to measure the flow properties of PP/PA-6 blends over the whole concentration range. Strong negative deviation was observed in the viscosity (η) versus volume fraction (ϕ) dependence at all shear rates and the lowest viscosity was reported at 25 wt % of PA-6. In a patent, PP/PA-6 blend was studied by Hinooka[57] where PP-g-maleic anhydride was used as a compatibilizer and observed that miscibility was improved along with the bonding strength. Mitsui-Petro-Chemical Industries Ltd.,[58] has developed an application of PP/PA-6 blends as a laminated plastic for plastic bottles. PP-grafted with acrylic acid, methacrylic acid and/or glycidyl methacrylate was used as a binder. The phase morphology and melt rheology at PP/PA-6 blends has been discussed in terms of its impact on the structure development during fiber spinning[59]. At higher temperature, phase coarsening was observed specially at PP 25 to 50 wt %. Reduction of the drop size with increasing shear rate during capillary flow was also reported. The molecular orientation in each phase was determined by wide-angle x-ray scattering. The results showed that orientation in PP was independent on composition, while reverse was true for PA-6. Goi, et al.[60], prepared blends with compositions 46.5 part PP, 65 part PA-6 and 3.5-part PP-g-acrylic acid and spun at 300°C temperature, which showed good spinability. The tensile properties of PP/PA-6/ ionomer were studied as a function of blend composition, method of blend preparation and draw ratio[61,62]. The modulus as well as the stress and strain at yield for blends containing 10 and 30 wt % of PA-6 was reported to go through a maximum at draw ratio $\approx$ 2:3, while for the compatibilized blends, the same properties showed a steady increase upto draw ratio $\approx$7.0. For the blend PP/PA-6/modified PP, Sato, et al.[63], reported that the tensile strength and modulus increased monotonically with increasing PA-6 content, while the tensile strength at break showed a large negative deviation. The effect of PA-6 concentration on the morphology and performance of PP-fibers was studied by Gorf, et al.[64]. Use of very low concentration of PA-6 resulted in a good fiber formation with better property. The blends with higher PA-6 concentration were found to have lower melt strength. Addition of maleated-PP to the blend was reported to significantly improve drawability and fiber performance. The effect of blend ratio of PP/PA-6 and PP/EPR on the mechanical properties of their binary and ternary blends was investigated by Shulun, et al.[65]. They analyzed the toughening mechanism for polypropylene. Evolution of the effect of level of functionality of maleated-PP as a compatibilizer on the reactive extrusion of PP and PA-6 and /or PA-6,6 was studied by Flaris and Boocock[66]. They have optimized the amount of maleation which is required for the compatibilization and for the elimination of delamination. The compatibilization mechanism of PP-g-itaconic acid as a compatibilizer for PA-6 rich PP blends was studied by Jingwei, and Wu[67]. The effect of functionalized PP on thermal, mechanical and morphological properties of PP/PA-6 blends was studied by Sathe, et al.[68]. According to them PP-g-butyl acrylate copolymer having a butyl acrylate content of 2.85 (wt %) acts as an interfacial agent for PP and nylon-6 blends. The grafted copolymer iPP-g-acrylic acid was investigated as a compatibilizer for blends of iPP and PA-6 and was reported to improve the blend compatibility[69]. The morphological and rheological properties of PP/PA-6 blends were investigated by Elliott and Townsend[70]. Maleated-PP was used as a compatibilizer. Braun and Schmitt,[71] have used PP-g-m-iso propyl-2-3-di-methyl benzyl iso cyanate(TMI) graft copolymer as a compatibilizer. Effect of recycled poly(vinyl butyral) film on the morphology and mechanical properties of the blends containing PP/PA-6 was studied by Lee and Soonja[72]. Use of di-butyl-maleate treated polypropylene (PP-g-DBM) as a compatibilizer under melt extrusion process was studied by Duxin and Demin[73]. They observed that improved compatibility increases the adhesion of the interfaces, decreases the particle size of dispersed phases and improves the mechanical properties. They have also observed that the compatibilization of the blend was affected by the grafting ratio of the compatibilizer but was not affected by the residue of PP-g-DBM. Maleic anhydride modified-PP copolymers with block structure was synthesized and used in the development of PP/PA-6 blends[74]. Morphological and mechanical properties of PP/PA-1010 blend were studied by Gao, et al.[75], where PP-g-

GMA was used as a compatibilizer. The compatibility and the mechanical properties of these blends were observed to improve with the addition of PP-g-GMA compatibilizer.

The dielectric, mechanical and morphological properties of PA-6/PP blend were studied by Yu et al.[76]. The influence of hyperbranched polymer grafted polypropylene (PP-g-HBP) on the interfacial adhesion between fusion bonded bilayers of PP and PA-6 was studied by Jannerfelat et al.[77]. Fiber formation in PP/nylon and PE/nylon binary compatibilized ternary blends in the region of phase inversion was studied by Tsebrekbo et al.[78]. The process of incorporation of a third polymer (5 - 30%) into binary (40:60, 50:50, 60:40) blends lead to formation of multiple microfibers with 3.2 - 5.0 mkm diam. They observed that compatibilizer can improve the compatibility and increase the kinetic stability of the melt and decrease the interfacial tension. The role of ethylene-vinyl acetate (EVA) as a compatibilizer is attributed to dipole-dipole interaction with amide bond of polyamide.

The effect of rubber functionality on microstructures and fracture toughness of impact – modified nylon-6.6/PP blends was studied by Wang and Mai[79]. They analysed the toughening mechanism in blends containing 60% nylon-66, 20% PP and 20% styrene-ethylene / butylene-styrene (SEBS) grafted with different levels of maleic anhydride (MAH). Blend of PP/PA-6 compatibilized with PP modified with MAH/vinyltriethoxysilane (PP-g-MAH and PP-g-VTES) and ethylene propylene rubber (EPR) modified with MAH, EPR-g-MAH) were investigated by Machtigall et al.[80]. The products were characterized by SEM, DSC, MFI and for mechanical properties. Li et al.[81] studied the morphology of PP/PA-6 blends compatibilized with PP-g-MHA. The graft copolymer was observed to improve the compatibility of PP/PA-6 blend effectively. SEM and TEM results indicated that the homogeneity of compatibilized PP/PA-6 blend was improved and the particle size of dispersed phase had significantly reduced. The effect of type of nylon on the compatibilization of PP/PP-g-GMA/PA-6 blends was studied by Tedesco et al.[82]. They have observed that in the compatibilized blends, a better dispersion of nylon in the PP matrix was attained resulting in better mechanical properties.

1.4.3 PP/PC blends

Due to the polar and non-polar nature of PC and PP, large interfacial tension results into coarser phase morphology, hence this type of blends have received limited attention in literature. Debkowski,[83] has studied the PP/PC blends. According to him addition of about 5 wt % of PP to PC reduces melt viscosity and improves processibility as well as the impact strength and modulus, reducing notch sensitivity and mould shrinkage. While, addition of about 10 wt % PC to PP improves it's processability and product appearance, as well as increases hardness, modulus and impact strength, reduces notch sensitivity and mould shrinkage. GE-plastic[84,85] reported that addition of an acrylic polymer enabled an increase of PP content that led to better gasoline resistance. Addition of styrene-(ethylene/butadiene)-styrene triblock copolymer(SEBS) allowed the use of upto 40 wt % of PC in blends for production of packing films[86]. The size and size distribution in PP/PC blends was studied in the full range of composition by Favis and Chalifoux[87]. They have reported that the degree at dispersion depended mainly on viscosity ratio (η), particularly when the high viscosity polymer formed the matrix. Both the particle size and the polydispersity increased with the minor phase content. The phase inversion region was affected by η as well as by the relative elasticity of the components. In the later studies the effect of processing on morphology was investigated[88]. Large differences in morphology were reported between the center and the outside region of the extruded strand, indicating the dominant role of the die zone for structure formation. Elongated fibers were observed to be at the rim while mostly droplets were seen in the center. In patent[89], blends which contain 20 to 80 (wt %) of either PP or PC compatibilized by a mixture of ethylene vinyl alcohol (EVAl) and SEBS grafted with either maleic anhydride (MAH) or furmaric acid are described. While, in an another patent[90], excellent mechanical properties, i.e., impact strength, heat distortion temperature, moldability and chemical resistance have been claimed with different compatibilizing systems, including SEBS, EPDM-g-styrene-acrylonitrile copolymer modified PP, polystyrene containing exazoline moieties, polybutylene terphthalate, etc. A preparation of PP/PC/PP triblock copolymer and its use as compatibilizer in PP/PC blends has been patented by Dow Chemical Co.[91] Fisa, et al.[92], have investigated the effect of processing on morphology and mechanical properties of PP/PC blends containing 0 to 40; wt % PC. Inspite of poor inter phase adhesion, enhancement of the modulus and strength was observed upto 40 wt % addition of PC. The maximum strain at break as well as the weld-line strength in injection moulded specimens was observed to decrease sharply. Good impact properties and high temperature performance of cycloalkylene-bisphenol polycarbonate and PP blends was claimed by Bayer[93]. Blends containing 4.0 % graft or block copolymers of PP-g-styrene, PP-g-styrene/acrylonitrile, PP-g-styrene/n-butyl methacrylate, PP-g-n-butyl methacrylate and SEBS as a compatibilizer were studied by Liang and Williams[94]. The effects of this compatibilizer on the crystallization, relaxation behaviour and mechanical properties was studied and significant improvement of blend performance was reported. Addition of PC to PP leads to enhancement of PP crystallinity and small increase in T_C. They found that compatibilization is limited to the amorphous phase. The relaxation behavior of the blend was found to be additive, calculable from those of the two major components, with compatibiliser playing only a small role. The heat deflection temperature, modulus, yield and fracture strength for blends containing PC (25 wt %) showed a negative deviation from the additive rule. Addition of a compatibiliser clearly improved the blend performance and PP-g-styrene acrylonitrile copolymer could work as a best compatibilizer. Electrical properties of blendmers of PP/PP-g-MAH/PC have been studied by Matsuura and Takamatsu[95]. The blendmer was used as an electorate air filter material.

Mechanical and morphological modification of PC (Mkrnlon-2805) with PP (0.5-5%) was studied by Barth[96]. The impact resistance was observed to increase by factor 5 with addition of 3% of PP. The influence of temperature on the interfacial tension for PP/PC and PA/HDPE blends was investigated by Chapleau et al.[97]. Here, interfacial tension was measured using the breaking thread method. Mitsubishi Eng. Plastic[98] developed a thermoplastic material based on PP/PC blend with good rigidity, impact strength, heat resistance and good processability. While development of rheological characteristics of fly ash filled PP/PC blend was studied by Chand et al.[99]. Single screw extruder was used to develop this blend and rheological study was carried out on MCR capillary rheometer. The effect of fly ash concentration on shear stress – shear rate, melt viscosity, shear rigidity, recoverable shear strain and first normal stress difference data were determined. This study shows that the melt viscosity increases and elasticity decreases, while recoverable shear strain reduces and shear rigidity increases with the increase of fly ash concentration.

1.4.4 PP/PET blends

Under this title blends with thermoplastic polyester PEST have been discussed. In spite of the early patents and the present activities there is no commercial PP/PET blend on the market. Using single screw extruder, flow properties for PP with polyethylene terepthalate were studied by Rudin et al.[100]. The blends were formulated within the low concentration region of dispersed phase : PP in PET and vice versa. Mutually beneficial effects are evidented. Two compositions of PET (50 and 70; wt %) showed lower viscosity. T_g for PET was reported to remain constant and dynamic modului were approximately additive. Blends of PET (40 to 20; wt %) were claimed to show a good balance of properties, making them suitable for manufacturing tubing, hoses, wire and cable jecketing etc.[101]. The blends of PP with poly-cyclohexylene dimethylene-co-ethylene terphthalate (PCTG) were also reported[102]. Impact properties were observed to increase with 30 to 50 wt %

of PP. Using PP-g-acrylic acid as a compatibilizer for PP/PET blend system Xantho and Yong[103] reported good dispersion and improved mechanical properties on compatibilization. In patent[104], carbonylated PP and EPR were used as a interfacial agent for PP/PET blends, which result into improved tensile properties. PP/PBT(polybutylterphthalate) blends with higher elongation at break and excellent impact resistance were reported by Kotlar, et. al.[105]. According to their process, PP was extruded with PBT and PP-g-GMA was used as a compatibilizer. Styrene was added during processing and was reported to inhibit the degradation during extrusion. Effect of processing parameters on *in situ* compatibilization of PP/PBT blends by one step reactive extrusion was studied by Hu and Lamba[106]. Where PP-g-GMA was used as compatibilizer. Results of the blends formulation showed that elongation at break and impact strength increased virtually linearly with decreasing feed rate (Q) or screw speed (N). The reactive compatibilization of PP/PET blends was also studied by Champagne and Peyrel,[107] using PP-g-GMA as a compatibilizer. Morphological and mechanical properties were studied. It was observed that mechanical behaviour changes from fragile to ductile region, while SEM observation showed that in compabilized region the PP average domain size decreases.

Preparation and properties of PP/PET blends were studied by Ebrahimian et al.[108]. In this paper PP/PET blends have been studied to find out the optimum ratio to give the best properties blends containing 5 to 50 wt % of PET. The morphological, mechanical and thermal properties were investigated. PP-g-MAH was used as a compatibilizer. A biaxially oriented laminated sheet from PP/PET blends for image receiving paper for printer was developed by Mitsubishi Polyester Film Corp.[109]. The effect of inclusions and blending on mechanical performance of recycled multi layer PP/PET/SiOx blends was studied by Wyser et al.[110]. Maleated PP was used as a compatibilizer and they have observed that at low compatibilizer concentration the blend exhibits a coarse morphology and brittle character. They have also observed that 5% compatibilizer shows fine morphology with excellent ductility, beyond this concentration a brittle interphase was formed.

Extrusion foaming of PET/PP blend with / without compatibilizer was studied by Wan et al.[111], while an excellent scratch-resistance in PP/PET blend for washing unit application was developed by Fujimura, et al.[112].

1.5 Proposed work

iPP being commodity semi-crystalline thermoplastic offers useful balance of heat and chemical resistance, good mechanical and electrical properties, and ease of processing. Whereas ABS is a versatile family of thermoplastics that contains three monomeric constituents acrylonitrile, butadiene and styrene. Each of these major components bring a different set of useful properties to the ABS.

Thus, resulting iPP/ABS blends can have the properties of ABS (toughness and modulus) and iPP, which makes the blend more cost effective, and with improved chemical resistivity and tensile properties. However, iPP/ABS are immiscible with each other and hence for better performance require a compatibilizer. With the aim of development of compatibilised iPP/ABS blends work was carried out as follows :

- Synthesis of various graft copolymers as compatibilizer.
- Characterization of the graft copolymers by FTIR, TGA, DSC and gravimetry.
- Use of selected graft copolymers (iPP-g-methacrylic acid and iPP-g-STY-MMA) in the preparation of iPP/ABS blends. Development of binary and ternary blends of different compositions.
- Determination of mechanical and morphological properties of the blend.
- Development of the mathematical models for the prediction of tensile modulus of the blends.

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