

Morphology and Mechanical Properties Investigation of Binary Polymer Blends Based on PP/SEBS and PP/PC

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HIGHLIGHTS

- By increasing PC in PP/PC binary blend, matrix tend to encapsulation of large PC drops.
- Particle size in PP/SEBS samples don't change considerably results from slight coagulation of drops together due to good SEBS interaction with PP matrix.
- PC particles in PP/PC blends can act as nucleation agent in PP crystallinity.
- All binary samples based on PP/SEBS exhibited a remarkable rise in the impact strength compared to pure PP which is attributed to the toughening effect of SEBS phase
- In PP/PC blends, the presence of large rod like composite droplets negatively affects the impact strength.

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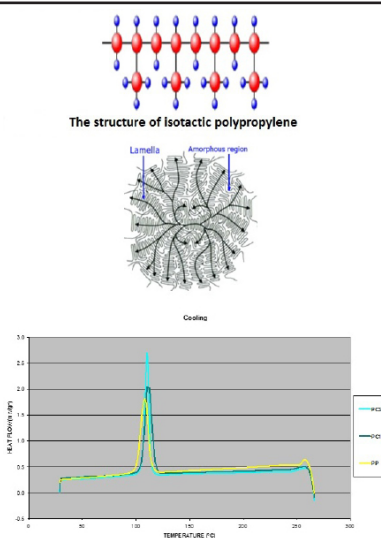
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ABSTRACT

In this study, eight binary blends of polypropylene (PP)/ poly [styrene-b-(ethylene-co butylene)-b-styrene] tri-bl^ck copolymer (SEBS) and polypropylene (PP)/ polycarbonate (PC) were produced at different composition through twin screw extruder. The composition of the minor phases was changed to correlate the morphology and mechanical properties of binary blends (PP/PC and PP/SEBS) through thermal properties. To serve this purpose and to study the composition weight, composition weight ratio of SEBS and PC was change from 10% to 30% in PP/PC and PP/SEBS. With increasing SEBS wt%, toughness grows slightly and in 30wt% of SEBS reached to its maximum value, which uniform morphology created by PP/SEBS plays important role in this matter. On the other hand, with increasing PC wt%, tensile properties have upward trend, but size of holes, due to lack of proper adhesion between two polymeric surface, rose. The results of DCS have shown that SEBS cannot act as nucleation agents (heterogeneous nucleation) but the PC particles in this situation can act as nucleation agent in PP crystallinity. Increasing SEBS percentages leads to immobility of PP segments and hamper the crystallinity. PC particles in PP matrix are heterogeneous nucleation agent, which leads to increasing in crystallinity temperature compared to the pure PP.

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1. Introduction

1.1. *Effective factors in formation and morphology control of the two phase polymeric systems Surface tension (the effect of adding compatibilizer)*

The final dimension of disperse phase stems from equilibrium between gravity and surface tension forces. In some researches done by scientists show that if adhesion is appropriate between phases (or low surface tension), Drops can sustain large deformation before fracture, which, in turns, leads to smaller drops morphology. Liang and co-workers [1] have worked on LLDPE/PVC by three compatibilizer. The relation between particle size and surface tension has been studied. They observed the dependence on particle size and surface tension in all three situations which is independence of compatibilizer yield. Meanwhile, they mentioned that the relation between surface tension and particle size can be predicted by Taylor theory as well. As a matter of fact, compatibilizer can affect deformation and drops fracture by decreasing in surface tension and consequently, deducing hydrodynamic stress which break up droplets with specific size. When a copolymer is used as a compatibilizer, by preventing coagulation leads to modifying and developing of morphology in comparison to drop fracture [2]. Lepers and co-workers [3] carried out a research on PS/EPR, they investigated the effect of surface tension and preventing coagulation by using two compatibilizer (symmetric and asymmetric block copolymer). Decreasing in surface tension in asymmetric was more than symmetric block copolymer and reduction in the particle coagulation by symmetric copolymer is more than asymmetric. Favis [4] used inomer as a compatibilizer in Polyamid/Polyolefin (PE and PP). He observed that in a specific concentration of compatibilizer, there is a sharp decline in particle size. This critical concentration depends on the disperse phase area of interface directly.

Zhang and co-workers [5] studied PP/PETG. They produced this blend 30/70 composition by SEBS, SBS and SIS compatibilizer in twin screw extruder. The micrographs represent that in this blend, PETG forms a fibril like structure for all blends and blends compatibilized by SBS have the smallest particles while SEBS blends have the largest particles. Moreover, compatibilized blends by SIS have average size between two mentioned systems (SBS and SEBS).

Nashar and co-workers [6] have studied LDPE/PET system. They used LDPE-g-AA and LDPE-g-AN as

compatibilizer. The blends contain LDPE-g-AA particles with smaller size and homogenous dispersion are compatibilizer. This is due to the interaction of these two compatibilizer with matrix. The blends contain LDPE-g-AA, acrylic acid groups (AA) make hydrogen bonds with PET chains while the connection between PET chains and LDPE-g-AA functional groups contributes to dipole interaction among nitrils and PET segments.

Fillipi and co-workers [7] have investigated the effect of different compatibilizer on LDPE/PA6. They used SEP-g-MA, SEBS-g-MA and HDPE-g-MA as compatibilizer. SEBS-g-MA and SEP-g-MA have dramatic impact on microstructure compared to HDPE-g-MA. Blends contain SEP-g-Ma and SEBS-g-Ma have smaller particles with better dispersion in comparison to blends with HDPE-g-MA as a compatibilizer.

Arefazar and co-workers have studied PET/PC and lantanon acetyl acetate as compatibilizer. Blends with compatibilizer show significant change regarding PC particle size and dispersion in PET matrix [8]. Arefazar and co-worker investigated PET/SBR and SBR-g-MAH as compatibilizer. They observed smaller particle size and better dispersion in PET matrix compared to the PET/SBR, which results in interaction between MA groups in SBR-g-MAH and carboxyl and hydroxyl groups in PET [9, 10].

1.2. *Viscosity Ratio*

Favis and Chalifoux [11] showed that in PP/PC even in approximately 13 torque ratio (disperse phase torque/ matrix phase torque), there is a significant deformation in disperse particle size in an internal mixer. They concluded that viscosity ratio play major role in morphology and the dimension of the dispersed phase. Furthermore, in 2 to 13 torque ratio, the dimension of dispersed phase increases to three to four times. More deduction in the dispersed particle in lower than 1 torque ratio can be obtained and the minimum particle size can be obtained in 0.25 torque ratio.

Favis and Therrein [12] demonstrated that in PP/PC, fracture of dispersed phase in high viscosity ratio in twin screw extruder with better efficiency happened in comparison to internal mixer and the relation between particle size and viscosity ratio in twin screw extruder is similar to the fracture of the Newtonian fluids in simple shear flow. Kim and co-workers [13] studied the effect of viscosity ratio in binary blend PP/EPR and ternary blend

PP/EPR/PE. The concluded that deformation of drops in matrix strongly accounts for viscosity ratio and to form fibrillar microstructure must be

$$\frac{\eta_d}{\eta_c} < 1$$

Hale and co-workers [14] investigated PBT/ABS and concluded that morphology in this blend strongly influenced by PBT melt viscosity.

1.3. Composition

The effect of composition on particle size in immiscible systems is of prime importance. In a research done by Li and co-workers [15], one of the systems was HDPE/PS which its morphology completely controlled by dispersed phase composition. So that in approximately 18%, 30% and 40% PS, co-continuous ratio is about 25%, 84% and 100% respectively. In 68% PS homogeneously destroyed and phase inversion happened. The dependence of phase structure to phase composition for two types of PP/EPM blends with different rheological behavior has been investigated by Fortenly and co-workers [16]. They showed that samples with similar viscosity of components compared to the samples with high difference viscosity ratio have more co-continuous structure for wide range of compositions.

Wang and co-workers [17] studied the effect of SEBS and SEBS-g-MAH composition on morphology and mechanical properties of PPO/SEBS and PPO/SEBS-g-Ma and observed that by increasing in the composition of SEBS and SEBS-g-Ma in both systems, morphology changes from sea-island in lower compositions to two continues phases in higher compositions. Morphology in both systems is under the influence of disperse phase composition.

1.4. Shear Stress

According to Taylor theory, the particle size of disperse phase has direct relationship to shear stress in mixing. Favis [18] mentioned that any changes in shear stress to two or three times, don't have any desire changes on morphology, which results from the fact that shear stress at interface due to slippage between layers and interfacial adhesion isn't continues. Moreover, in another study by Favis and Therein [12] on PP/PC in a twin screw extruder shows that shear stress doesn't have dramatic impact on

particle size.

1.5. The relation of microstructure and mechanical properties in polymer blends

Wong and co-workers [19, 20] studied PA66/PP modified by SEBS-g-MA in different MA composition. The reported that tensile strength and stiffness rise with increasing SEBS-g-MA in 75/25 of PA66 and PP respectively. Hence, in 50/50 of PA66/PP, with increasing of SEBS-g-MA opposite trend was observed which stems from phase inversion in this system (PP consider as matrix).

Wang and co-workers [21] used DGEBA as compatibilizer in PTT/MB in different weight composition (0 to 5 wt%). They have seen that with adding DGEBA, tensile properties such as tensile strength, Young Modulus and elongation at breaks have modified by increasing weight percentage of this compatibilizer. Moreover, impact resistance increased.

Gonzalez and co-workers [22] studied the influence of phenoxy on mechanical properties of PTT/Phenoxy. With increasing weight percentage, young modulus increases and shows a positive deviation from rule of mixture, so that yield stress rise with increasing in phenoxy composition. Impact resistance shows a negative deviation in similar percentage and rich of phenoxy. Impact resistance have downward trend at first and upward trend in the following. SEMs prove this result.

Yi and co-workers, PP/PET microfibrillar processed in a single screw extruder by slit mold. They investigated the effect of increasing of PP-g-GMA as compatibilizer on morphology. They observed that thanks to surface tension by compatibilizer, mechanical properties, tensile strength and impact resistance modified, however young modulus decrease due to lack of stiffness of compatibilizer compared to PET microfibriles.

Jiang and co-workers [24] studied the effect of epoxy resin in PP/SEBS-g-MA. They showed that all mechanical properties increase when cure agent add to epoxy resin during PP/SEBS-g-MA/epoxy processing in comparison to uncured epoxy. This effect is due to creation of specific microstructure surrounding SEBS-g-MA particles. In addition, there is a good balance regarding stiffness and toughness in this system.

Rastin et al. [25] studied a series of binary and ternary polymer blends based on HDPE, polyamide-6 (PA6), poly(ethylene-co-vinyl alcohol) (EVOH), maleated HDPE and suggested that the

core-shell morphology remains unchanged by addition of 0.5, 1.5, 3, and 6 wt.% of HDPE-g-MAH to the ternary blends. But analyzing of impact strength, showed that the addition of 1.5 wt.% HDPEg-MAH to 75/25 MHDPE/EVOH and MHDPE/PA-6 pairs increases about 2.5 and 6 times the impact strength with respect to their corresponding non compatibilized blends, respectively.

Saeb et al. [26] demonstrated that the mechanical properties of high density polyethylene (HDPE), poly (ethylene -co- vinyl alcohol)(EVOH), and polyamide-6 (PA6) binary and ternary blends are strongly influenced by processing temperature. In addition, it has been observed that the yield stress increase upon increasing temperature due to enhanced interfacial adhesion between HDPE/EVOH matrix and PA-6 dispersed phase.

Saeb et al. [27] described that for ternary polymer blends based on HDPE/PA-6/EVOH with PA-6 and EVOH as minor components with different weight ratios, yielding behaviour of prepared ternary blends is dependent on the minor component fraction possibly due to the formation of voids at the interface of polymers.

In another research, the morphology of polypropylene (PP)/poly (trimethylene terephthalate) (PTT)/poly (styrene-b-(ethylene-co-butylene)-b-styrene) (SEBS) 70/15/15 ternary blends prepared by twin screw extrusion was investigated as a function of maleic anhydride-assisted SEBS compatibilizer composition on the morphology and mechanical properties by Moini, et al. [28] It is found that by diminishing SEBS to SEBS-g-MAH weight ratio, the morphology changed from the pure core-shell to a multi-phase system consisting of both core-shell and detached particles. This transition in the phase morphology caused some advantages on the mechanical properties, so that the blend consisting of 50/50 weight ratio of SEBS/SEBS-g-MAH exhibited the maximum value of the impact strength.

Moini et al. [29] have shown that the morphology of the ternary PET/PC/NBR blend is influenced by NBR-g-GMA compatibilizer and by increasing the rubber content, the rod-like structures were disappeared; besides, toughness was increased. Furthermore, by increasing PC content, rod-like structures have seen by morphological study; however, core-shell droplets formed in the blend structure caused enhancing the impact strength and reducing Young's modulus.

In this article, the influence of the composition

of PC and SEBS on microstructure and mechanical properties of PP/PC and PP/SEBS through DSC will be investigated.

2. Experimental

2.1. Materials

The following materials were used in this work:

(i) An iso-tactic polypropylene homo-polymer (PP), SEETEC H5300 supplied by LG chemical company(Korea) (MFI: 3.5 g/10min, 230 °C, 2.16kg), (ii) Polycarbonate (PC), Makrolon 2858 purchased from Bayer Co(Germany)(MFI: 10 g/10min, 300 °C, 1.2kg), (iii) Poly(styrene-b-(ethylene-co-butylene)-b-styrene) (SEBS) tri-block copolymer, Kraton™ G1652 supplied by Shell Chemicals (29% styrene; molecular weight; styrene block 7000, EB block 37500)

2.2. Blend Preparation

In this study, 8 binary blends were produced at different weight ratio using Brabender co-rotating twin screw extruder (diameter of screw = 2 cm, length/diameter ratio = 40).The various compositions used for this research are reported in Table1. Prior to processing, the compositions were dried in an oven for at least 17h at 80°C. The barrel of extruder has six temperature-control zones and their temperatures were set at 230-235-240-245-250-255°C (from hopper to die). The screw speed was maintained at 130 rpm.

Table1: Various compositions of binary polymer blends

Code No.	PP(%wt)	PC(%wt)	SEBS (%wt)
PC10	90	10	0
PC15	85	15	0
PC20	80	20	0
PC30	70	30	0
SE10	90	0	10
SE15	85	0	15
SE20	80	0	20
SE30	70	0	30

2.3. Mechanical Properties

After melt blending of designed compounds in twin-screw extruder, the blends were quenched in cooling water bath and pelletized in a granulator. Dried blends were molded to form tensile and impact specimens using an ENGEL injection molding machine. The Barrel temperature profile was 180 °C (hopper) to

240 °C (nozzle) and the mold temperature was maintained at 40 °C. Tensile stress-strain data were obtained using Galdabini testing machine in the rate of 50 mm/min according to the ASTM D-638. Moreover Izod impact strength was done for notched specimens according to ASTM D-256 using Zwick pendulum-type tester.

2.4. Morphological Studies

In order to evaluate the effect of particle size and the type of resulted morphology on the mechanical properties of PP/PC and PP/SEBS binary blends, scanning electron microscopy (SEM) micrographs were obtained using AIS-2100 scanning electron microscopy supplied by SERON Company through fracture surface of impact specimens. Before doing scanning electron microscopy, the impact samples were fractured in liquid nitrogen and consequently were etched by cyclohexane for 24h to remove SEBS minor phase. Then, the etched samples were gold sputtered to make the samples conductive.

2.5. Thermal Properties

This test leads to better estimate of compatibility of component in blends. This device (200F3Maia) was made in NETZCH Germany. Approximate weight of samples were 7.5 mg and heating rate was 10 °C/min. Three heating-cooling-heating cycles from 30 to 265 °C were selected. The percent of crystallinity was calculated using following equation:

$$X_c = \frac{\Delta H_f}{\Delta H_{f^0}} \times \frac{1}{\omega_{pp}} \times 100$$

In this equation:

ΔH_f is enthalpy PP

ΔH_{f^0} is enthalpy isotactic PP (100% crystal) equal to 209 J/gr for PP[31]

ω_{pp} is weight fraction of PP in the blend

3. Results and Discussions

3.1. Morphology investigation of binary blends PP/PC and PP/SEBS

Morphology of PC10, PC15, PC20 and PC 30 belong to PP/PC and SE10, SE15, SE20 and SE 30 belong to PP/SEBS according to the table 2 are in fig. 1. Furthermore, the results of the SEM analysis present in table 3.

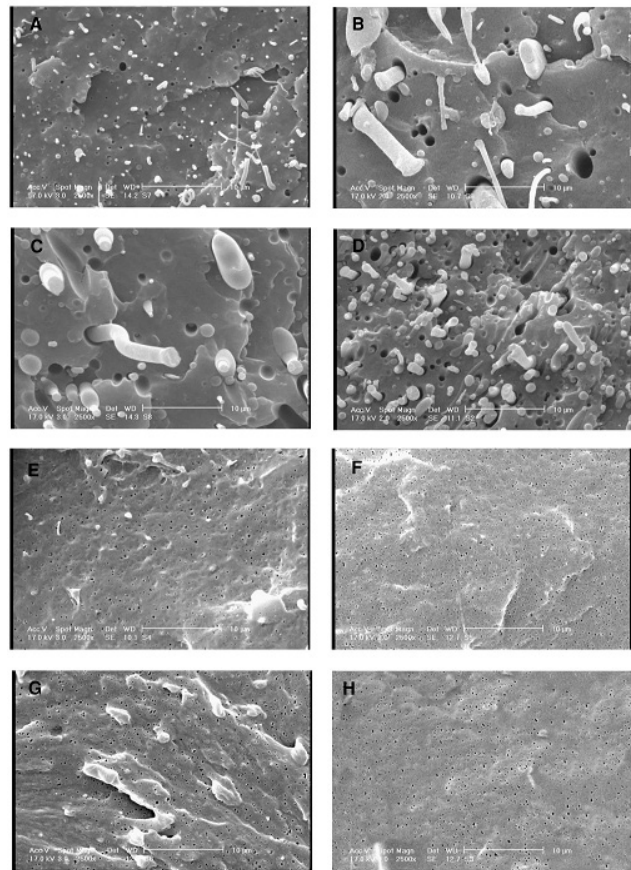


Fig1. SEM micrographs of PP/PC and PP/SEBS at different compositions according to table1.

Relying on SEMs and their analysis, in PP/PC by increasing PC composition to 20% dimension of spherical drops of PC in PP matrix increase due to agglomeration. Moreover, the size of cavities increased due to lack of proper surface adhesion. On the other hand, by increasing PC in this blend, matrix tend to encapsulation of large PC drops, which, in turn, is completely obvious in 15%, 20% and 30% as well as Rod PCs either independence or surrounded exist in that weight fraction. In all samples, hallow cavities implying high interfacial tension can be seen.

PC30 has the highest surrounded drops (either spherical or rod). PP/TPE in separated phases is similar to modified polymeric blends by rubber. Additionally, PP matrix and thermoplastic elastomer has influence on size, particle, shape and its distribution which leads to various morphologies formation especially in high compositions [30]. Referring to SEMs of PP/SEBS it can be inferred that uniform distribution of SEBS drops like black holes which demonstrate drops were extracted by cyclohexane solvent in PP matrix. This is due to the fact that good miscibility of PP with drops owing to Block Ethylene-Butylene

Table2. Results of Image analysis of SEM micrographs according to table1.

Number of rod like PC particles	Number average size of rod like PC particles (μm)	Number of individual rod like PC particles	Number average size of individual rod like PC particles (μm)	Number of composite droplets	Number average size of composite droplets (μm)	Number of individual PC core particles	Number average size of individual PC core particles	Number of individual SEBS particles	Number average size of individual SEBS particles (μm)	Data type
7	0.67514	16	0.40413			50	0.40536	46	0.32439	PC10
9	1.79655	7	1.26314	7	1.05842	20	0.74055	19	0.93258	PC15
6	1.7955			12	1.40008	15	1.17993	12	1.479	PC20
17	1.13282	11	0.87327	19	1.12438	52	0.87062	27	0.69581	PC30
								130	0.2076	SE10
								139	0.15342	SE15
								155	0.21248	SE20
								225	0.1309	SE30

Table3. DSC results of the PP/SEBS and PP/PC binary blend samples

DSC					
XC (%)	wpp	ΔHf (J/gr)	Tm ($^{\circ}\text{C}$)	Tc ($^{\circ}\text{C}$)	Sample code
36.68	1	76.67	167.3	108.3	PP
48.79	0.85	86.67	164.766	110.162	PC15
53.55	0.7	78.34	163.738	110.244	PC30
47.003	0.85	83.5	167.128	110.218	SE15
54.45	0.7	79.66	164.768	111.202	SE30

in SEBS tends to PP matrix. With increasing SEBS, the number of SEBS drops increase. Particle size in these samples don't change considerably results from slight coagulation of drops together due to good SEBS interaction with PP matrix, which leads to better stress transfer from matrix to disperse phase and better fracture of drops in matrix and slight coagulation.

3.2. Thermal Properties

According to the table 3, SEBS cannot act as an effective core in PP crystallinity (heterogeneous nucleation), which leads to increasing crystal temperature of PP in both SE15 and SE30. Hence, due to miscibility of PP with SEBS, the high probability of EB block penetration towards PP chains and creation of micelles because of aggregation results in ineffectiveness of SEBS as a nucleation agent in PP crystallinity. On the other hand, due to miscibility and interaction SEBS with PP, the possibility of creation of incomplete crystalline structure increased, which, in turns, leads to low melting temperature of pure PP and this fall increase by increasing in SEBS wt%. Another considerable point is that with increasing SEBS wt% from 15 to 30, heat of fusion declined, which

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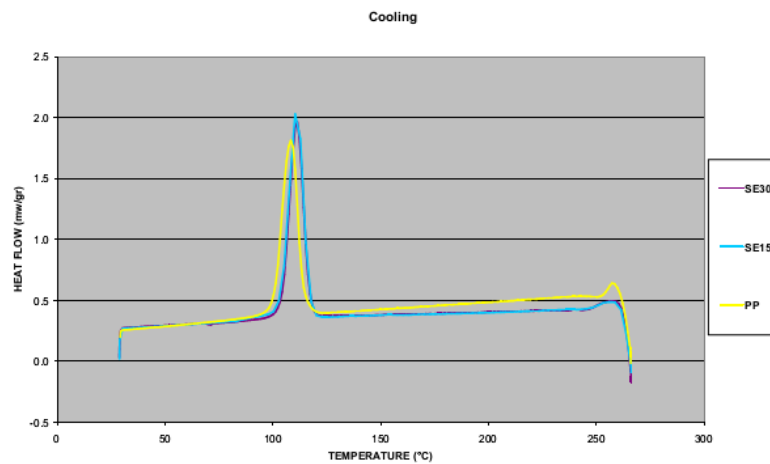
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which results from better interaction between components with increasing SEBS percentage, which leads to immobility of PP segments and hamper the crystallinity. On contrary, with increasing SEBS, degree of crystallinity decreased. Therefore, degree of crystallinity increased compared to the pure PP. Figure 2 represents the cooling cycle of SE15, SE30 and pure PP. Based on figure, with adding SEBS to PP, peak goes higher and narrower, which results in speeding up the crystallization. The results of table 4 demonstrates that PC particles in PP matrix are heterogeneous nucleation agent, which leads to increasing in crystallinity temperature compared to the pure PP [31]. Another point is that more particle and distribution in matrix, due to nucleation, more PP crystallinity. This trend is completely clear in the table. On contrary, heat of fusion of PC15 and PC 30 rise in comparison to pure PP, which results from increasing in PP crystallinity. Diagram 3 illustrates the cooling cycle of PC15, PC30 and pure PP. Re-

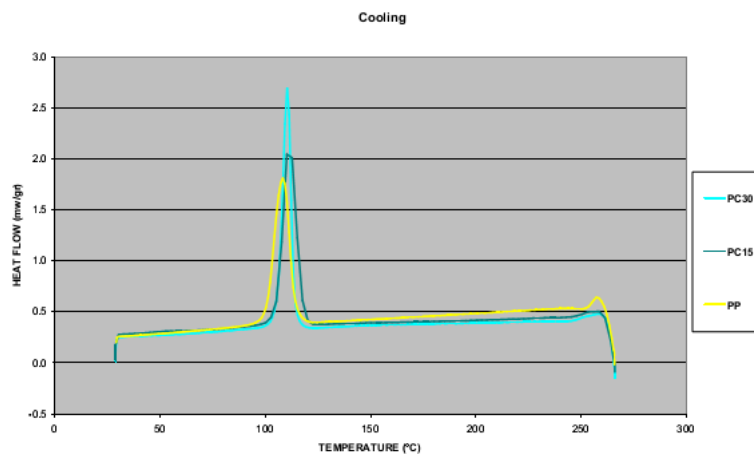
lying on this diagram, with increasing from PC to PP, peak goes higher and narrower, which results in increasing in the rate of PP crystallinity specially in 30wt%. Therefore, PC particles in this situation can act as nucleation agent in PP crystallinity.

3.3. Investigating Mechanical Properties

Table 4 shows the mechanical properties of PP/SEBS and PP/PC. According to this table, it is clear that with increasing SEBS in all PP/SEBS samples, yield stress, young modulus and tensile strength decrease due to elastomeric nature, but elongation at break, regarding elastomeric properties, increases. Due to proper interaction and adhesion between PP and SEBS with increasing in PP, impact resistance increase. This rise in SE30, with the highest SEBS percentage, reaches to its maximum value. In accordance with the PP/PC mechanical properties, it can be inferred that with increasing in PC wt%, tensile



.Fig. 2. Diagram SE15 and SE30 samples cooling cycle



.Fig. 3. Diagram PC15 and PC30 samples cooling cycle

strength increase slightly and yield stress compared to pure PP rises resulting from the nature of high stiffness of PC. Decreasing in yield stress from PC10 to PC30 is a consequence of existence of more structures which matrix surrounding bigger PC particles. In this condition, micro cracks grow in interface causing weakening interface [32]. Since stiffness nature of PC (its high yield stress), with increasing in PC weight percentage leads to rise in yield stress despite lack of interfacial adhesion. Elongation at breaks in PC10 shows a fall which is sharper in higher percentage resulting from weak interaction in these weight percentage is the results of surrounding of bigger PC particles leading to the growth of cracks in interface. SEMs confirms above result easily. Since the stiffness of PC, young modulus increases, reaching to its maximum value in PC30, arise from the presence of more rod like PC individual and surrounded structure in this sample. Tensile strength shows a decline at first arising from undesirable interaction and adhesion between PP and PC, however it slightly increases from PC20. Despite due to an increased in composition and consequently agglomeration in the system the nature of PC stiffness resolves this effect and tensile strength increased. Regarding impact resistance, adding PC to PP matrix due to lack surface interaction to transfer stress, leads to declining in impact resistance of PP. The intensity of this drop from PC10 to PC 15 is more than other samples resulting from surrounded PC particles by PP matrix which is more in higher percentage and causing growth of micro cracks at interface. On the contrary, presence of rod like structures in this system can cause better impact resistance attributing to better orientation of this structures which are perpendicular to crake growth. Therefore, referring to the abovementioned remarks, it can be imply that first factor is much more effective

than second factor which is presence of rod like structure and plays a major role.

4. Conclusions

Investigating the effect of disperse phase on morphology and mechanical properties of ternary blends from two binary PP/PC and PP/SEBS blends leads to these results: PP/PC with a heterogeneous morphology stemming from undesirable adhesion between PC and PP at interface. Therefore, disperse phase plays key role in increasing stiffness in this blend. Moreover, PP/SEBS with a desirable morphology wherein SEBS plays important role in increasing toughness in this blend. Finally, DSCs prove that SEBS cannot act as nucleation agents (heterogeneous nucleation) but the PC particles in this situation can act as nucleation agent in PP crystallinity.

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Table 4. Mechanical properties of the PP/SEBS and PP/PC binary systems.

Impact strength (J/m)		Tensile strength (MPa)		Young modulus (MPa)		Elongation at break (%)		Yield stress (MPa)		Properties
S.D	average	S.D	average	S.D	average	S.D	average	S.D	average	Samples
0.47	25.66	1.43	19.44	22.60	1167.40	19.96	279.17	0.39	32.30	PP
0.42	24.90	1.79	16.20	9.43	1387.64	3.37	107.23	0.35	34.17	PC 10
0.14	20.00	0.98	12.92	103.49	1402.25	23.38	101.35	0.06	33.99	PC 15
1.68	19.03	0.70	23.86	55.33	1437.72	2.79	29.99	0.10	36.30	PC 20
0.14	17.60	1.29	28.12	0.57	1749.51	4.50	29.03	1.41	37.30	PC 30
5.05	65.13	1.12	18.15	5.01	1081.07	185.04	629.03	0.36	29.04	SE 10
23.33	171.50	0.69	19.03	18.89	978.61	85.80	380.25	0.64	27.25	SE 15
7.94	317.00	0.08	17.06	16.21	965.54	136.72	688.54	0.29	26.18	SE 20
13.58	551.33	0.37	16.70	1.26	898.40	11.72	823.28	0.13	23.15	SE 30

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