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Mechanical Properties Thermoplastic Laminates of Polycarbonate-Polyetherimide Blend with Glass Fibers

Singh JC[1]*, Srivastava M[2], Singh RK[2] and Yadav SB[3]

[1]Indian Institute of Technology, New Delhi, India
[2]Central Institute of Plastics Engineering and Technology, Hajipur, India
[3]Defence Material & Stores Research & Development Establishment, Kanpur, India

Abstract

Thermoplastic Laminates of Polycarbonate (PC)/Polyetherimide (PEI) blend with glass fibers were prepared by solution blending of PC & PEI in different ratio of 100/0, 95/5, 90/10, 85/15 and 80/20 using Dichloromethane as solvent followed by applying the resulting blend solutions over the cut sheet size (30 × 25 cm) of woven mat glass fibers & dried overnight, then stacked up 12 flies of the impregnates and consolidated in a hot press at 300°C and 3-4 ton (30 kg/cm^2) pressure for a duration of one hour & cooled. The laminates of PC/PEI blends/woven mat glass fibers were studied for their mechanical properties viz: Tensile, Flexural, Impact and Inter Laminar Shear Strength and thermo-mechanical properties viz: TGA, DSC, DMA as well as examination of morphology by SEM. The mechanical properties of laminates of PC/PEI blends/woven mat glass fibers shows an increasing trend in Inter Laminar Shear Strength, Tensile and Flexural Strength upto 20% PEI while Impact Strength shows an increase upto 10% and decreases while the percentage of PEI increases. Glass transition temperature of the laminates increases with increasing content of PEI and the degradation kinetics of TGA reveal that the decomposition temperature decreases with increasing content of PEI. DMA of laminates shows that the thermo-mechanical properties get improved at a higher temperature with the presence of PEI.

Keywords: Polymer composites; Polycarbonate; Polyetherimide; Glass fibers; Mechanical properties

Introduction

Composite materials are engineering materials with two or more material constituents having significantly different physical and chemical property, which remain separate at the macroscopic level in the finished structure with their high stiffness, high strength, and low weight properties [1]. Polymer matrix composite (PMC) has been used for many applications. PMC is the type of composite with polymer material acting as matrix and has another material fiber or more in it. PMC has established themselves as engineering structural materials, not just as laboratory curiosities or cheap stuff for making chairs and tables. This came about because of not only the introduction of high performance such as glass, carbon, boron and aramid but also some new and improved engineering materials [2].

Glass, carbon and Kevlar fibers are extensively being used as reinforcement in variety of forms, such as continuous roving, woven roving and fabrics, and random chopped strands, all meeting particular processing needs [3], in the resin matrices like polycarbonate(PC) polyetherimide (PEI), polyether-ether-ketone (PEEK), unsaturated polyesters, epoxies, phenol-formaldehyde etc. The composites of these, with an epoxy resin such as glass epoxy, and carbon epoxy composites are most widely used material. Polymer composites offer several advantages over conventional materials like, higher fatigue strength, corrosion resistance, very high strength to weight ratio, ease of fabrication, assembly, installation etc. Further, they are very economical when manufactured using appropriate fabrication technique.

Composites are composed of reinforcement, resins, fillers & additives. Each of these constituents & materials of ingredient plays an important role in the processing & final performance of the product. The resin or polymer is the 'glue' that holds the composite together & influences the physical properties of the product. The reinforcement provides the mechanical strength. The fillers & additives are used as processor performance aids to impart special properties of the product.

The mechanical properties & composition of fiber reinforced polymer composites can be tailored for their intended use. The types & quantity of materials selected in addition to manufacturing process to fabricate the product, will affect the mechanical properties & performance, important consideration for the design of composite products include: types of fiber reinforcement; percentage of fiber or fiber volume; orientation of fiber (0, 90, 45 or a combination of these); types of resin; cost of product; volume of production; manufacturing process; service conditions etc.

However, selection of matrix has a major influence on the inter-laminar shear properties of the composite material. The inter-laminar shear is an important design consideration for structures under torsion loads [4]. The mechanical properties of resins used for matrix materials in composites must consider the effect of reinforcement material. Domination of composite properties by the reinforcement is true for many properties such as tensile strength, flexural strength, and thermal expansion [5]. The molecule in these polymers contains rigid aromatic rings that give them a relatively high glass transition temperature 'Tg' and excellent dimensional stability at elevated temperature [6,7].

Polymer matrix composites are currently being used in a wide range of products to suit various industrial requirements, because of their design flexibility, high performance characteristics, low potential costs and energy saving. These materials are thus used in various fields which include agriculture, chemical, electrical, electronics,

***Corresponding author:** Singh JC, PHD Pursing, Indian Institute of Technology, Hauz Khas, New Delhi, India, Tel: +91-7827958920; E-mail: chandrajeettz@gmail.com

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biochemical, energy conservation and aerospace sectors, military, chemical industries include reactors, pressure vessels, piping, plating baths, fan blades etc. [8].

The blends of PC/PEI exhibit a higher heat distortion temperature and an improved flexural strength and tensile strength over the polycarbonate component alone and have higher impact strength than that associated with the polyetherimide component of the blends. In addition, the blends may exhibit good flame resistance. Multilayer composites comprising polyetherimide layers adjacent polycarbonate or co-polyester carbonate layers prepared by lamination of separate layers extruded in the normal fashion, or more typically by co-extrusion through a die head whereby the layers become intradielaminated, have the benefits of a rigid, high modulus, impact resisting first layer in combination with a high temperature resistance, high modulus at high temperature, solvent resistant, abrasion resistant and low coefficient of friction second layer [9]. In this line of activities, an attempt was made to prepare a typical engineering thermoplastic blend laminates by blending PC/PEI through solution blending technique & impregnating the polymer blend solution with the woven glass fibre mat and stacked the impregnated woven glass fabric mat and then pressed in a hot press followed by cooling to get the laminates which were tested & evaluated for thermo-mechanical & morphological properties [10,11].

Materials and Methods

The polycarbonates (PC) (Lexan XHT1141) with density of 1.2 g/cm^3 & melt volume rate of 16.0 cm^3/10 min. and the Polyetherimide (PEI) (Ultem1010) with 1.27 g/cm^3 & melt flow rate of 17.8 g/10 min was obtained from M/S.SABIC Innovative Plastics, India; the Woven glass Fibers (E) mat was obtained from Decan Glass Fibers, India and the Dichloromethane (DCM) LR was Ltd. Varodara, India obtained from Samir Tech-Chem Pvt.

Polymer blending

Polyetherimide (PEI) pellets are first put in a hot air oven for 1 hour at 120°C so that moisture present in the PEI get evaporated, the then different weight of (5 gm, 10 gm, 15 gm and 20 gm) is dissolved in dichloromethane (DCM) solvent. The PEI gets dissolved normally in 5-6 hour. The PEI solution was mixed with PC in the following compositions given in Table 1 and mixed thoroughly by using mechanical stirrer at 20°C for 1 hour. Then each blend solution was heated at 500 for 15 minutes so that the traces of solvent get evaporated and results in a fairly consistent fluid in-order to apply conveniently to the woven mat glass fiber.

Laminate preparation

The cut sheet size (30 × 25 cm) of woven mat glass fibers was impregnated with the blend solutions & dried overnight, then stacked up 12 flies of it together in between the Teflon sheets and consolidated in a hot press at 300°C and 3-4 ton (30 kg/cm^2) pressure for a duration of one hour & cooled.

Experimental techniques

Mechanical properties: The laminates were examined for various parameters as given in Table 2 and subjected to testing for different mechanical properties. The Tensile properties such as tensile strength, Tensile modulus, strain at failure & poisson's ratio as per ASTM D3039, Flexural properties such as flexural strength & flexural modulus as per ASTM D790 & Inter-Laminar Shear Strength as per ASTM D2344 using a Tinius-Olsen (T-O) 10,000- pound Universal Testing Machine

Sample Code	PC (%)	PEI (%)	PC (gm)	PEI (gm)	DCM (ml)
B0	100	0	58.6	0	120
B1	95	5	55.67	2.93	120
B2	90	10	52.74	5.86	120
B3	85	15	49.81	8.79	120
B4	80	20	46.88	11.72	120

Table 1: Composition of PC-PEI blends.

Laminates Code	Weight of laminates (gm)	Thickness of laminates (mm)	Fiber weight fraction in the laminates (gm)
C0	267.0	4.00	0.8125
C1	270.5	4.00	0.8166
C2	267.3	4.00	0.8209
C3	274.8	4.00	0.8235
C4	280.1	4.00	0.8250

Table 2: Details of Laminates of PC/PEI with glass fibers.

USA, testing machine and Impact strength as per ASTM D256 using Instrumented Instron Software of flat laminates were tested.

Thermogravimetric analysis (TGA): The thermal characteristics of the laminates were determined by Thermogravimetric analysis. The experiments were carried out in the N2 atmosphere using Universal V4.2E, TGA 2950 TA Instruments, and the USA. The sample weighing, 5 mg & heating rate, 10°C /min. was used and thermograms were obtained as a plot of residual weight against temperature. The sample was heated from room temperature up to 1000°C. The percent residual weight was calculated at 900°C.

Differential scanning calorimetry (DSC): Differential scanning calorimetry, Universal V3.9A, TA Instruments (Model 2910), USA was used for thermal analysis of laminates. The samples of about 5 mg were placed in aluminium's pans with pierced lid. The tests were conducted under a nitrogen atmosphere at a heating rate of 10°C/min.

Scanning electron microscopy (SEM): The phase morphologies of laminates including the fractured samples after impact & flexural tests were observed using Carl Zeiss EVO-50 XVp low vacuum SEM. The tensile fractured samples were sputter coated with a thin layer of gold palladium alloy prior to SEM examination.

Dynamic mechanical analysis (DMA): Dynamic mechanical analyser, Universal V3.9A, TA Instruments (Model 2980), USA was used for analysing viscoelasticity characteristics of the laminates. The dynamic mechanical analysis is a well-known method for determining viscoelastic properties by applying a controlled sinusoidal strain to a sample and measuring the resulting stress. The measurements were carried out at a heating rate of 10°C/min from ambient to 350°C at a fixed frequency level of 1 Hz. The samples for the test were rectangular bars.

Results and Discussion

Tensile properties

Tensile Strength values for different laminates by increasing percentage of modifier PEI with PC in the matrix is shown in the Figure 1A and B. It can be concluded that the tensile strength of the laminates increases as percentage increases of the weight of modifier PEI. From the graph it also concludes that the tensile strength values for 5 percent PEI weight ratio laminate and 10 per cent PEI weight ratio laminate come nearly same. From Tensile Modulus graph it was found that the tensile modulus of the laminates slightly increases by increasing the %

weight ratio of PEI with PC and at 20% PEI weight ratio with PC Tensile Modulus value is highest. The best tensile strength is observed in case of (PC: PEI/80:20) blends because there is much better miscibility than (PC: PEI/90:10) (Table 3).

Flexural properties

It was found that the flexural strength of the laminate is increased with increasing the PEI content up to 20% in the PC-PEI blend composition. It was found that the flexural modulus of laminates also increases than unmodified resin laminate. From the graph of flexural strength *vs.* Percentage of PEI for laminate it was found that the flexural strength of the blends are increasing as we increase the PEI content in the blend up to 20% PEI in the PC resin. The flexural strength for this composition increases to about 25.95 times greater than the unmodified resin (Figure 2A-B and Table 4).

Impact properties

It was found that the impact strength of the laminates is decreased with increasing the PEI content in the blend up to 20% in the PC resin. From the graph of impact strength *vs.* Percentage of PEI for laminate it was found that the impact Strength of blends gradually decreases up to composition 20% PEI. Therefore the Impact Strength of the laminates decreases on increasing PC i.e. increasing PEI weight ratio in PEI-PC matrix (Figure 3 and Table 5).

Inter-Laminar shear strength

ILSS refers to the shear strength between the two plies of the laminate. ILSS depends primarily on the matrix properties and fiber/matrix interface shear strength rather than the fiber properties. ILSS of PC laminates with PEI interface is higher than the unmodified PC resin. From the graph of inter-laminar shear strength *vs.* different percentage of PEI for laminate. It indicates that by the use of PEI the adhesion property of the resin can be increased as we increase the PEI content in the matrix laminates (Figure 4 and Table 6).

Thermo gravimetric analysis

It is concluded from the above line chart that the decomposition temperature (T_{max}) decrease as we increase the percentage of modifier PEI content, although it is better for the PEI content 5% and 10%, at this temperature the max weight loss occurs in the samples. From the above graph it can be concluded that the residual weight slightly

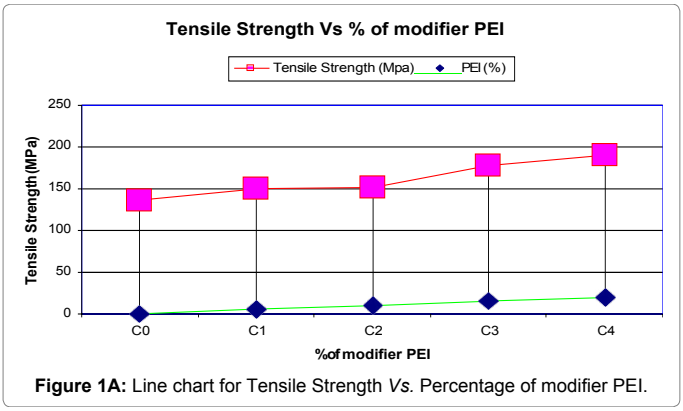

Figure 1A: Line chart for Tensile Strength *Vs.* Percentage of modifier PEI.

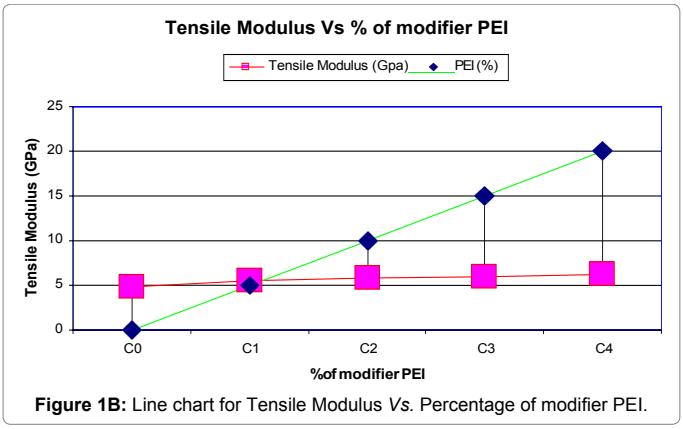

Figure 1B: Line chart for Tensile Modulus *Vs.* Percentage of modifier PEI.

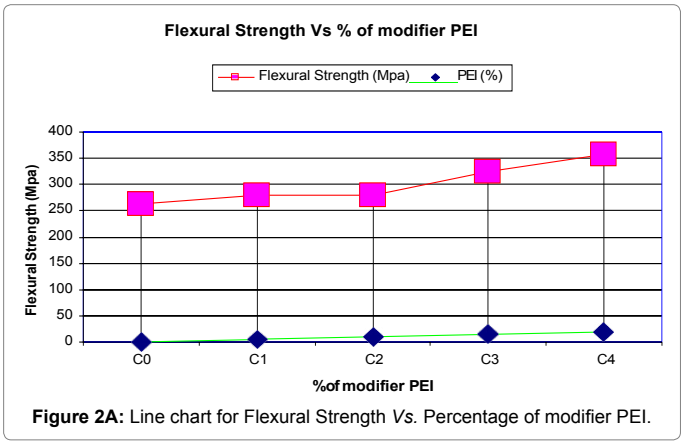

Figure 2A: Line chart for Flexural Strength *Vs.* Percentage of modifier PEI.

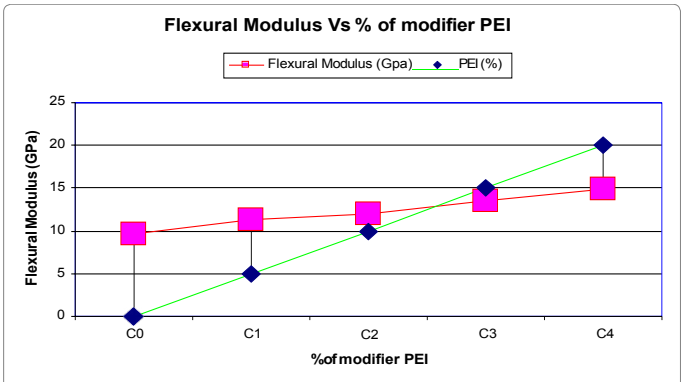

Figure 2B: Line chart for Flexural Modulus Vs Percentage of modifier PEI.

Laminates Code	Tensile Strength (MPa)	Tensile Modulus (GPa)
C0	135.59	4.86
C1	149.32	5.56
C2	151.73	5.85
C3	177.97	5.95
C4	190.82	6.25

Table 3: Tensile Properties of Laminates.

Laminates Code	Flexural Strength (MPa)	Flexural Modulus (GPa)
C0	263.39	9.67
C1	279.75	11.28
C2	280.16	12.01
C3	325.31	13.56
C4	357.93	14.87

Table 4: Flexural Properties of Laminates.

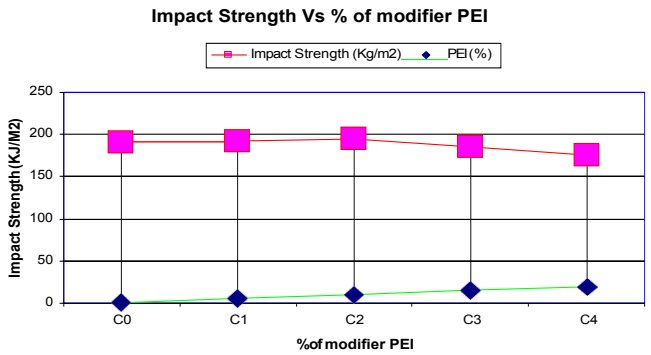

Figure 3: Line chart for Impact Strength Vs Percentage of modifier PEI.

Laminates Code	Impact Strength (KJ/M2)
C0	190.46
C1	175.25
C2	169.17
C3	161.78
C4	153.61

Table 5: Impact Properties of Laminates.

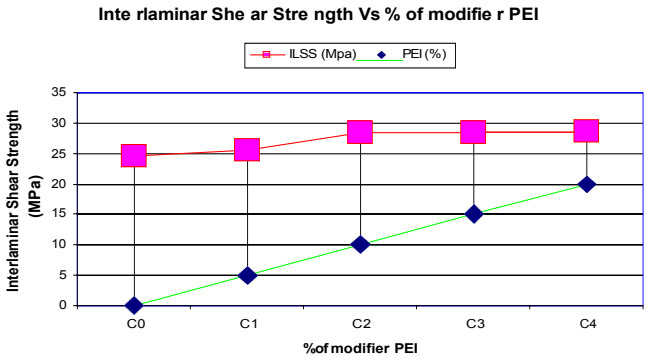

Figure 4: Line chart for Inter-Laminar Shear Strength Vs Percentage of modifier PEI.

Laminates Code	Inter laminar Shear Strength (MPa)
C0	24.60
C1	25.50
C2	28.40
C3	28.50
C4	28.60

Table 6: Inter Laminar Shear Strength (ILSS) of Laminates.

decreases with increasing the % of modifier PEI up to 10% & again rise up with increases the % of modifier PEI then decreased suddenly for the composition of PC/PEI: 80: 20 system. It is found from the above table that the residual weight is found the maximum for the composition PC/PEI: 85/15 that means it is best used for higher temperature applications.

Differential scanning calorimetry

From the above graph of maximum temperature *vs*. percentage of PEI it is indicated 1st & 2nd Tg (°C) are found the maximum for the PC/PEI: 90/10 & minimum for the composition of 15 wt. percentage of PEI. The literature shows that the blend of PC & PEI are immiscible but my work show miscibility up to 20% weight ratio of PEI in PC-PEI blends system (Figure 5A-B and Table 7).

Dynamic mechanical analysis

In general all the modified systems have more value of storage modulus at room temperature than a neat one. From the graph of temperature of storage modulus *vs*. Percentage of PEI shows that the storage modulus of PC/PEI (85:15) was found maximum at room temperature. It means this composition have more resistance towards deformation by an applied force thus possess high stiffness. It was found that the Onset temperature in general increases with increasing the content of PEI in the system than neat one and maximum onset temperature was found for PC: PEI (80:20) matrix composition. From the graph of inflection temperature *vs*. Percentage of PEI shows that the inflection temperature is almost same for all composition of the matrix. The maximum inflection temperature is shown by the ratio of matrix composition (90:10). From the graph of end temperature *vs*. % of PEI

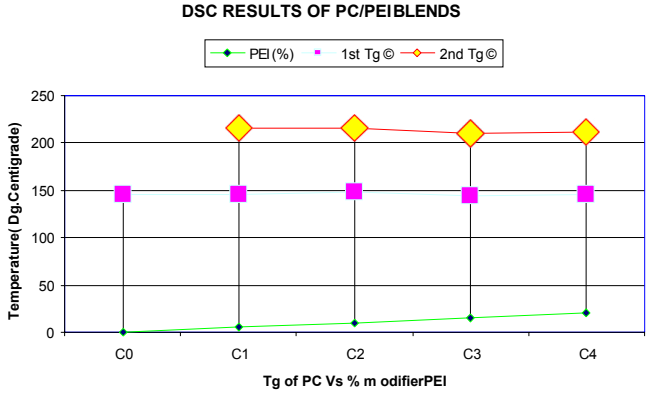

Figure 5A: Line chart for Glass Transition Temp. Vs Percentage of modifier PEI

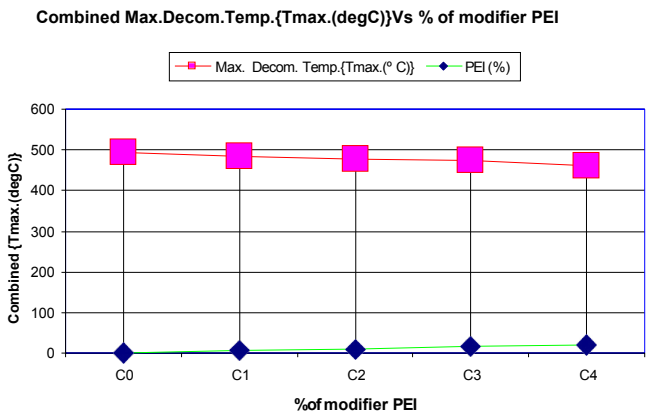

Figure 5B: Line chart for Comb.Max.Decom. Temp. (Tmax.°C) Vs Percentage of modifier PEI.

Testing Code	% of PC (by wt.)	% of PEI (by wt.)	1st Tg (°C)	2nd Tg (°C)
C0	100	0	145	0
C1	95	5	146	216
C2	90	10	148	216
C3	85	15	144	210
C4	80	20	145	212

Table 7: DSC data's of neat Polycarbonate and its Blends with Polyetherimide.

shows that the end temperature of the laminates shows fluctuation. The end temperature increases upto the ratio of matrix composition (90:10) then sudden decreases for the PC/PEI (85:15) composition after this sharply increases for the composition of PC/PEI (85:15). From the above graph, the maximum end temperature is shown maximum for the composition (90:10). It means this system can resist applied force without complete loss of storage modulus (stiffness) (Figure 6A-6G and Table 8A-B).

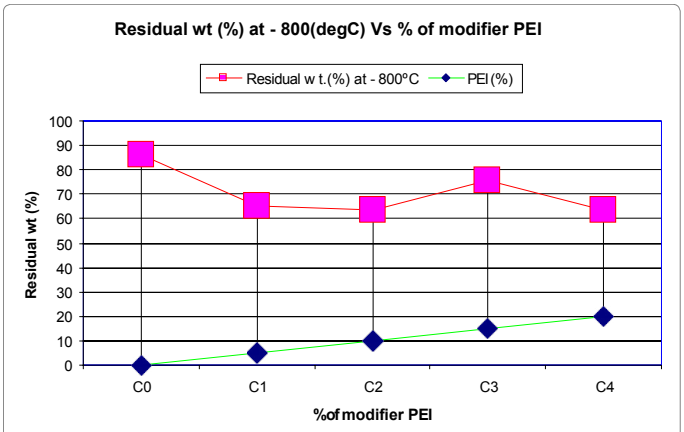

Figure 6A: Line chart for Residual Wt (%) Vs Percentage of modifier PEI.

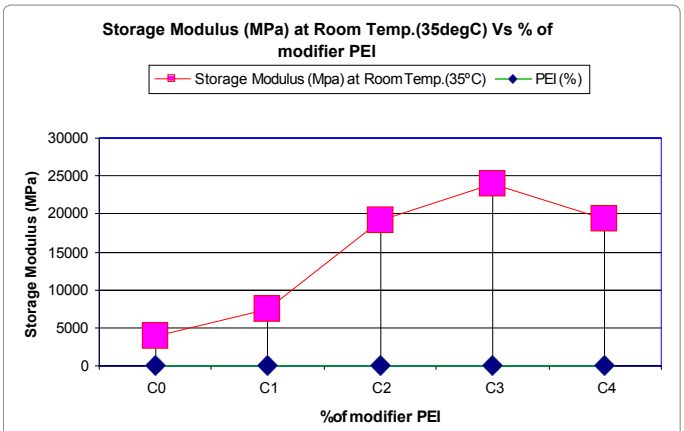

Figure 6B: Line chart for Storage Modulus (MPa) at Room Temp. (35°) Vs Percentage of modifier PEI.

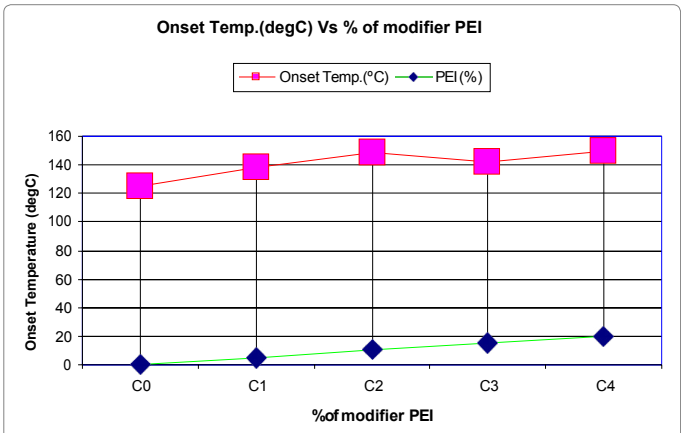

Figure 6C: Line chart for Onset Temp. (°C) Vs Percentage of modifier PEI.

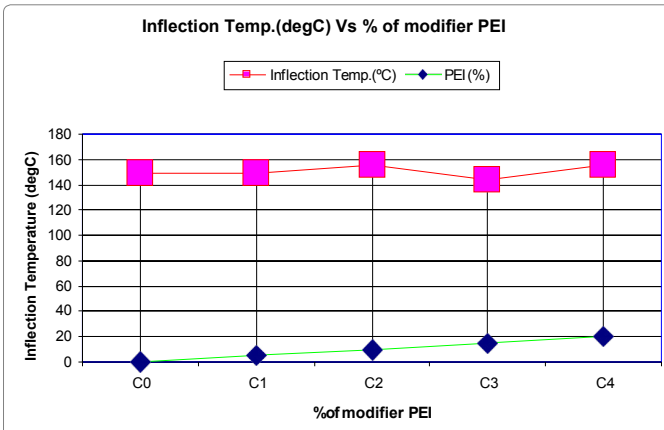

Figure 6D: Line chart for Inflection Temp. (°C) Vs Percentage of modifier PEI

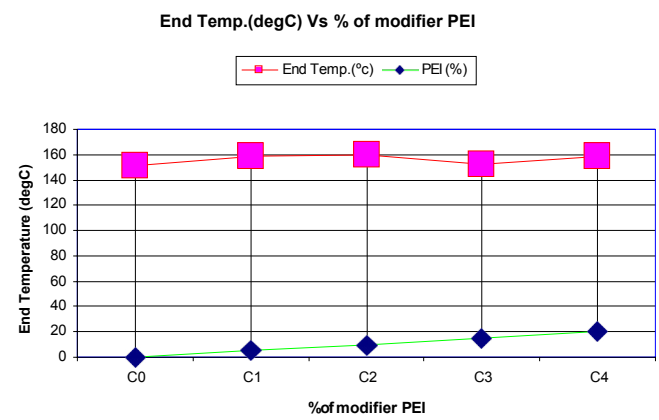

Figure 6E: Line chart for End Temp. (°C) Vs Percentage of modifier PEI.

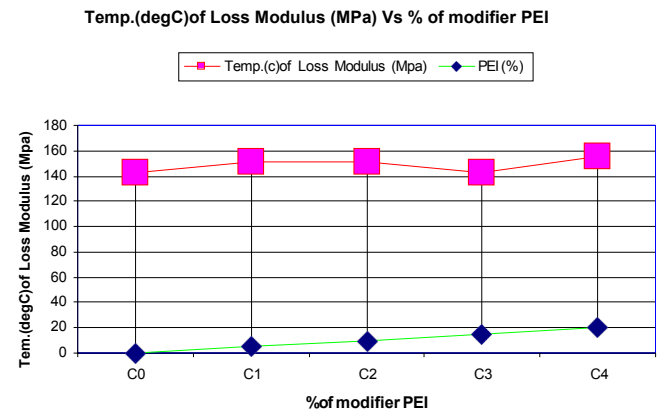

Figure 6F: Line chart for Temp. (°C) of Loss Modulus Vs Percentage of modifier PEI.

Morphological studies

The SEM micrographs were taken for the fractured surface of the neat PC and their blends with PEI. The different micrographs were taken for each specimen of the PC, PC/PEI blends at magnifications of 1000X, 500X, 300X, 1.00KX. From them we can confirm the occurrence of complete miscibility between the two components, showing all the surfaces as a single phase with similar ductile fractures, typical of thermoplastics.

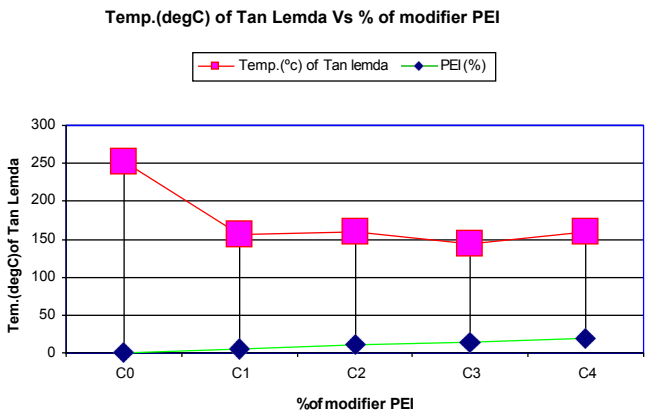

Figure 6G: Line chart for Temp. (ºC) of Tan δVs Percentage of modifier PEI.

Testing Code	% of PC (by wt.)	% of PEI (by wt.)	Max. Decom. Temp. (final) Tmax.(ºC)	Residual wt (%) at - 800 (ºC)
C0	100	0	495.42	85.94
C1	95	5	484.23	65.37
C2	90	10	478.15	63.63
C3	85	15	473.48	75.74
C4	80	20	460.69	63.45

Table 8A: TGA data's of neat Polycarbonate and its Laminate with Polyetherimide.

Testing Code	% of PC (by wt.)	% of PEI (by wt.)	Storage Modulus (MPa) at Room Temp.(35ºC)	Onset Temp. Temp.(ºC)	Inflection Temp.(ºC)	End Temp.(ºC)
C0	100	0	3960	125,00	148.89	150.75
C1	95	5	7450	138.44	149.54	158.95
C2	90	10	19200	148.27	155.45	159.50
C3	85	15	24000	142.30	142.30	152.54
C4	80	20	19400	149.80	149.80	158.75

Table 8B: DMA data's of neat Polycarbonate and its Laminate with Polyetherimide *Vs.* (Loss Modulus and Tan δ).

From SEM micrograph of Figure 7A-7O is observed that slightly phase separation occurs in case of PC/PEI (80:20) composition of the matrix.

It was observed that for the PEI content 10% & 15% SEM photograph presented as a rough fractured surface than neat PC and the crack propagation direction is not clearly defined specially for Figure 7. It was observed that there is no effective bonding between matrix and fiber that is clearly seen in Figure 7O. Here some impressions of glass fibers on the matrix are clearly shown (Figure 7A-7O).

Conclusion

From above study it was observed that the blends of PC with PEI resin influences the thermal and mechanical properties of that system by making the modified resin better in properties up to PEI content 20% in the system than a neat one. Increase in the glass transition temperature of the blend found as PEI content increases. As a theory of miscible polymer states that the glass transition temperature of the blend lays between the glass transitions temperatures of the components. The maximum glass transition temperature 210 ºC is for the 80/20 composition as it has maximum PEI component, which lies between the Tg of neat Polycarbonate and 20% of PEI Tg. Mechanical properties i.e. Tensile strength, Tensile Modulus, Flexural strength and Flexural modulus is maximum for the 80/20 (PC/PEI) FRP composite 190.82 ± 5 MPa, 6.25 ± 5 GPa, 357.93 ± 5 MPa and 14.87 ± 5 GPa respectively and minimum for the neat Polycarbonate/Polyetherimide (100/0) FRP Composite (135.59 MPa, 4.86 GPa, 263.39 MPa and 9.67 GPa respectively) respectively and increases as PEI content in PC/PEI matrix increases. Thermoplastic FRP composite is one of the most used composites in the field of defense and aerospace applications. The PC/PEI blends increase its mechanical properties almost 1.2-2 times

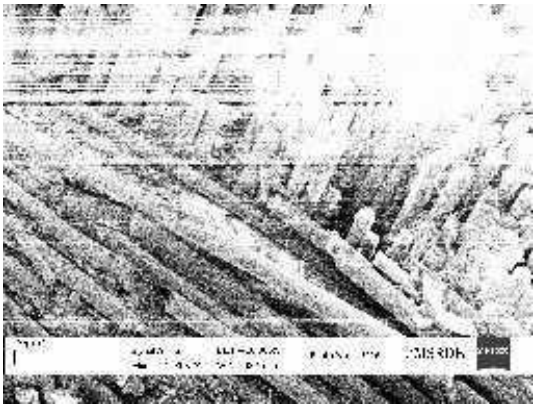

Figure 7A: 1000X Mag. PC Neat.

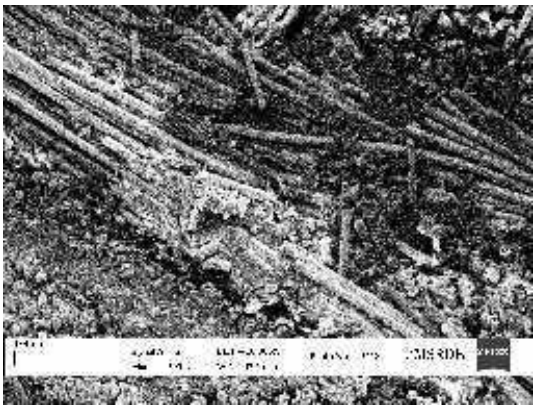

Figure 7B: 500X Mag. PC Neat.

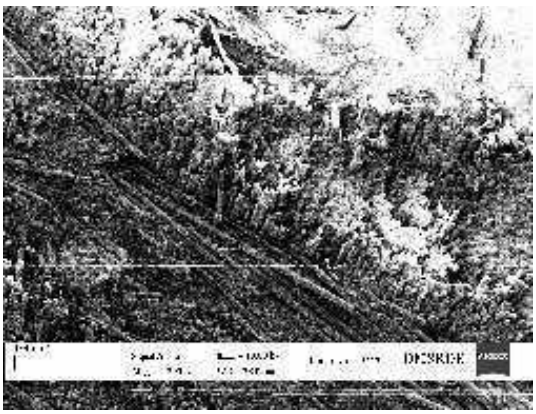

Figure 7C: 300X Mag. PC Neat.

Figure 7D: 1000X Mag. PC: PEI (95:5).

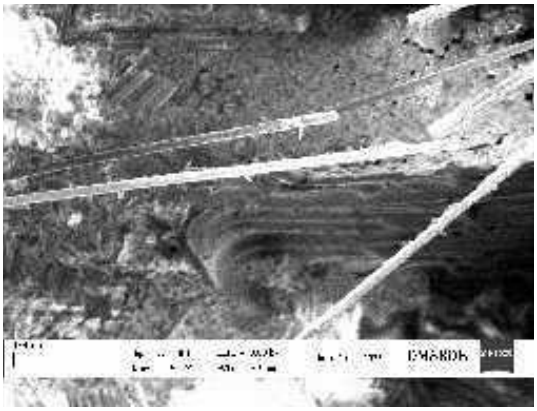

Figure 7E: 500X Mag. PC: PEI (95:5).

Figure 7F: 300X Mag. PC: PEI (95:5).

Figure 7G: 1000X Mag. PC: PEI (90:10).

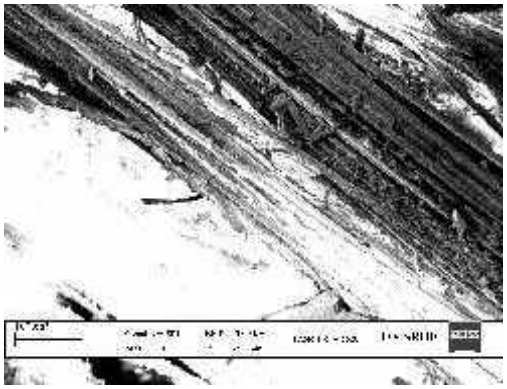

Figure 7H: 500X Mag. PC: PEI (90:10).

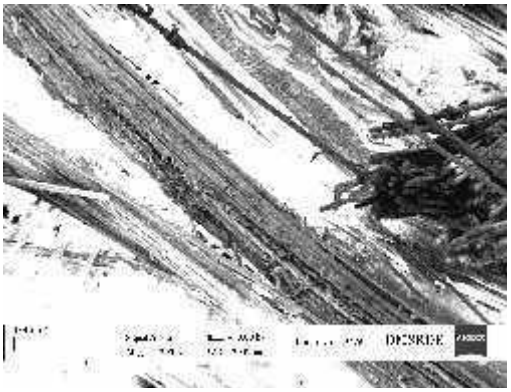

Figure 7I: 300X Mag. PC: PEI (90:10).

Figure 7J: 1000X Mag. PC: PEI (85:15).

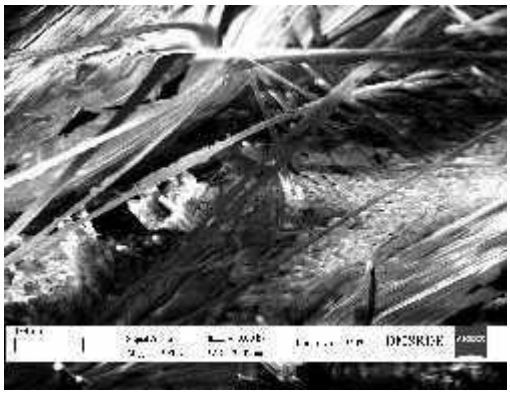

Figure 7K: 500X Mag. PC: PEI (85:15).

Figure 7L: 300X Mag. PC: PEI (85:15).

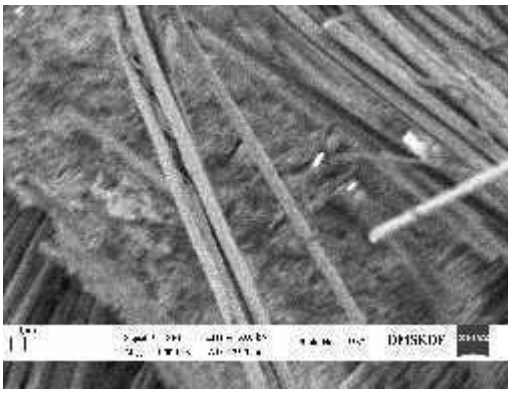

Figure 7M: 1000X Mag. PC: PEI (80:20).

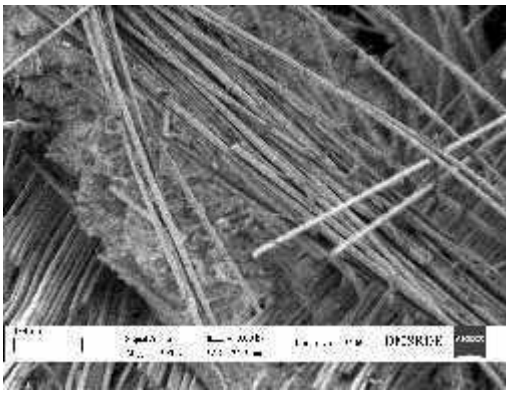

Figure 7N: 500X Mag. PC: PEI (80:20).

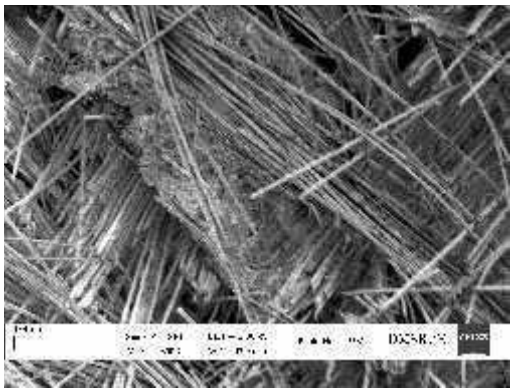

Figure 7O: 300X Mag. PC: PEI (80:20).

to the neat PC-FRP composite. The thermal properties also improved by the addition of PEI in the Polycarbonate. Hence instead of using neat Polycarbonate FRP composite it is beneficiary to using PEI blended Polycarbonate FRP composite for the defense and aerospace applications.

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