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Eco-friendly foam biocomposites based on cellulose extracted from date palm leaves and low-density polyethylene

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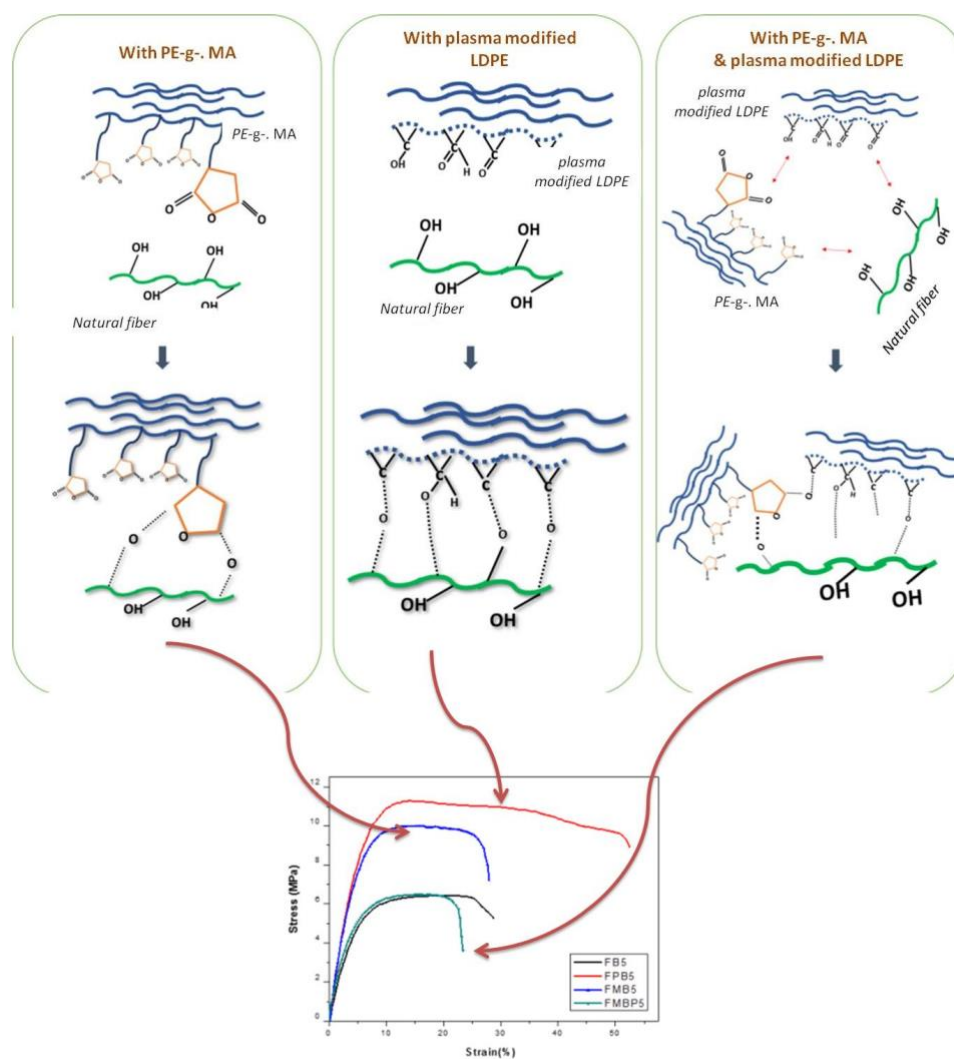
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Graphical Abstract:



Abstract: The design of composite materials using environmentally compatible materials and adopting sustainable preparation techniques are highly required for a better environment. The present study is a step towards developing an environment friendly alternative to foam-based composites. The aim of the study is to know the effect of natural fibers from date palm on the property of foam biocomposites. We adopted an environment friendly method of compatibilization between the polymer and the fibers for better reinforcement. The compatibility was done by environment-friendly physical modification using plasma treatment. Plasma treatment provides surface modification and this is an economical solution for further processing. In fact three types of interfaces have been created: 1) by the addition of maleic anhydride, 2) use of plasma treated LDPE, 3) the combination of maleic anhydride and plasma. The overall performance was the best by the use of plasma treatment alone. The effect was carefully analyzed based on the morphological, mechanical and thermal studies. Finally, morphology-property correlation was established

1. Introduction :

The awareness to protect our environment has brought out an overwhelming change in the research field in developing products based on environment friendly polymer materials [1-4]. The art of making polymer composites based on the requirement of the industry has brought out novel materials with excellent properties. Perceptively designed polymer composites possess unique physical, mechanical and thermal properties which are governed by the polymer matrix. Other major parameters that can change the final properties are the fillers and additives. Polymer foam composites are lightweight materials with many attractive properties such as low density, impact damping, thermal insulation, flexibility etc. Polymer foams have wide range of applications in offshore exploration of oil and gas, in electromagnetic shielding thermal and acoustic insulation [5]. The properties of polymer foams not only depend on the intrinsic properties of the polymer, but also on the foam morphology, such as cell density, cell size and size distribution [6]. It is important to improve the properties of the composite foams, which is still not yet explored completely. Addition of fibers (long or short) to composite foams provides modified microstructure with improved strength and damage tolerance. Fillers in micro or nano level can be used to prepare polymer nanocomposite for foams. One of the reasons for adding fillers while synthesising polymeric foam nanocomposites is to improve the strength of the foam without affecting its lightweight. Recent research focuses on adding micro and nanofillers so that they can act as

nucleation sites to decrease cell size along with reinforcing the composite. Lee *et al.*[7] have studied the foaming behaviours of wood fiber/high density polyethylene (HDPE) composites with small amounts of nanoclay. The results suggested that the addition of nanoclay improved the cell morphology of the wood fiber/HDPE composite foams as its content and degree of dispersion increased. The research in foam industry mainly aims at producing an environment friendly foam product which uses natural resources and non-hazardous materials. It is expected that these foams during processing can impart excellent properties to the final product. Natural fiber based biodegradable polymer foam composites are yet to be produced in high quantities in foam industry. Cellulose fibers have been foreseen to be used as the potential fillers in foam composites. There are many concerns relating to environment protection which leads to the development of biocomposites through environment friendly methods. The greater usage of natural fibers and the environmental friendly methods for foam preparation are a viable alternative to produce environmental friendly polymeric foams with comparable properties. Although the uses of natural fibers have successfully been used to prepare good composite materials with excellent properties, they are not widely used in designing foam materials. However, recently the use of natural fibers in foam composite also gained recognition due to the wide scope of product development using natural resources as well as property enhancement. Andre *et al.* [8] have studied new applications for foam composites of polyurethane and recycled butyl Rubber was supported in a polyurethane matrix generating two new products of distinct characteristics and properties [9-13]. Usually in polymer composites the foaming agents are blended with the polymer or added directly into the hopper and from there down through the barrel to the mold. Thermal decomposition of chemical reaction of the material may be either endothermic (heat absorbing) or exothermic (heat generating). Endothermic foaming agents primarily produce CO₂ while exothermic mostly generate N₂. The blowing agents like chlorofluorocarbons, (CFC's) hydrochlorofluorocarbons (HCFC's) hydrofluorocarbons (HFC's) are mostly not preferred because of their negative environmental impact on the ozone layer. These have high global warming potential although these are ideal blowing agents. Liquid CO₂ are also used as a blowing agent because it does not have the same environmental issues as CFC's and HCFC's. It has a low global warming potential and is inexpensive. However endothermic chemical blowing agents such as mixtures of sodium bicarbonate and citric acid are gaining in popularity for foaming thermoplastics because of properties such as: fast degassing times, improved cell structure, and endothermic decomposition and is also environment friendly.

The present study is a step toward developing a comparatively environment friendly alternative to foam-based composites usually prepared by using foaming agents like CFC's HCFC's and HFC's which are harmful to environment. For environmentally friendly blowing agents the by-products are not harmful. The blowing agents used were citric acid and sodium bicarbonate. The aims of the study are; (1) to have a deep understanding of the effect of natural fibers from date palm trees on foam biocomposite, (2) to adopt an environment friendly method of compatibilization between the polymer and the fibers for better reinforcement. *Considering the use of cellulose from agro waste and the introduction of eco-friendly blowing agents, we have adopted a process which is environmentally friendly.* Plasma treatment provides surface treatment and it is an economical solution for further processing. Plasma treated LDPE surface can modify the interfacial properties and thus improve ultimate composite characteristics. The method is economic also as the materials used for the preparation is not costly [14,15,16]. A lot of studies are reported that explain the significance of plasma treatment on polymers which shows the improved adhesion to natural fibers [17,18,19,20].

The current study employs low-density polyethylene (LDPE) as a resilient and non-brittle material. LDPE offers several advantages such as lightweight, low cost, flexibility (low modulus), toughness, chemical resistance and ease of sealability etc [21]. An environment-friendly procedure for compatibility between date palm fibers and LDPE has been adopted by plasma treatment (physical modification) of the LDPE surface. The work will pave the way in the value addition of date palm leaves which is widely available especially in the middle east.

2. Experimental

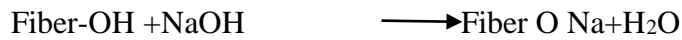
2.1. Materials and methods

The base polymer which was used for the preparation of foam composite was LDPE (grade Lotrène® FD0274) supplied by QAPCO with melt flow index 2.4g/10 min. The blowing agents used were citric acid and sodium bicarbonate supplied by Sigma Aldrich. The date palm leaves were collected locally from the Qatar University gardens.

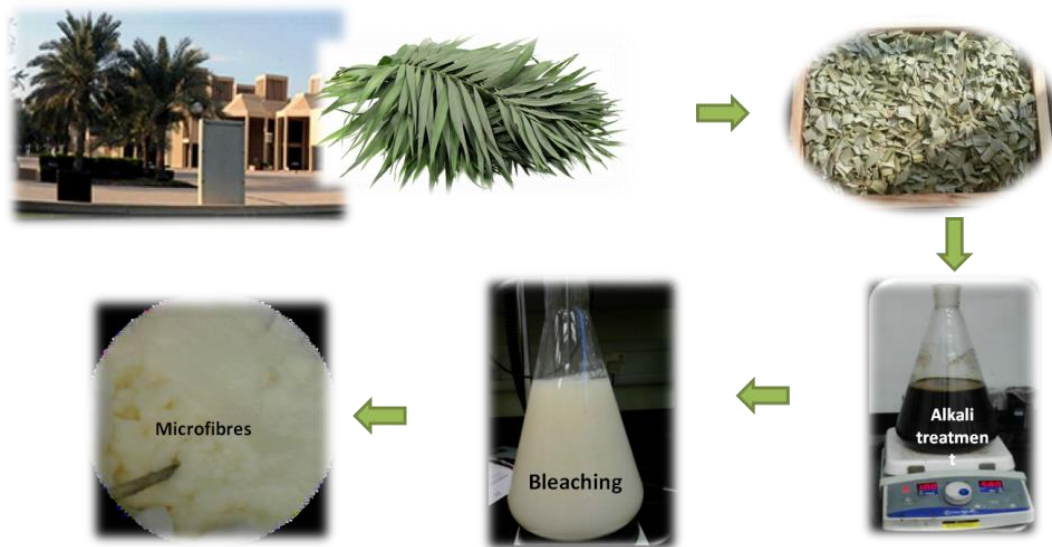
2.1.1. Preparation of microfibers from date palm leaves

The date palm leaves were washed well with water to remove the dirt and other pollutants. It was dried and was cut into small pieces of about 1cm length. The leaves were then dried in an oven for overnight. It was then ground into fine fibers using a blender. The powder was separated into 40µm, 60 µm and 100 µm particles using a micro sieve shaker. 45 g of

100µm powder was taken and was treated with 2%NaOH for 3 hrs at 90°C to remove the hemicellulose, lignin, and other substances in small amount like pectin bounded to the cellulose. The fibers were then washed with water till it became neutral.



The alkali treated fibers were then bleached with an equal volume of sodium hypochlorite (1:3) and buffer solution (NaOH and acetic acid).The process was repeated 5times to remove as lignin content, and the white fibers obtained after the bleaching was washed with distilled water several times. Finally, the fibers were dried for analysis.(Scheme 1)



Scheme 1: The microfibrils prepared from date palm leaves

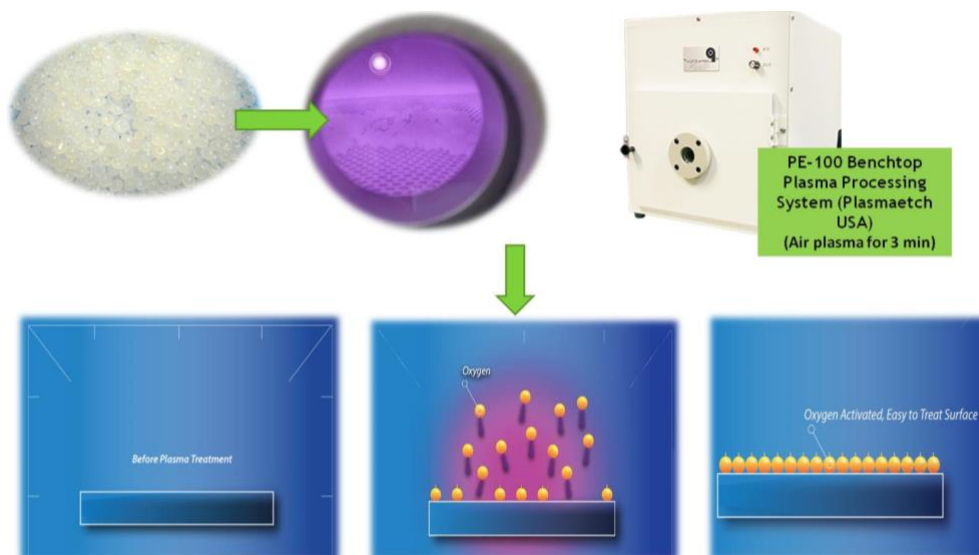
2.1.2. Preparation of polymer foam composite

2.1.2.1. Preparation of masterbatch

10g of the polymer was melted mixed in xylene and was mixed with fiber and maleic anhydride. It was then cast and xylene was removed by placing it in the oven at a temperature of 55°C for one week, after that it was dried and powdered.

2.1.2.2. Preparation of plasma modified LDPE(PLDPE) as a compatibilizer.

LDPE was treated with air plasma under the following condition: The LDPE pellets were put in the chamber and subjected to air plasma treatment. Prior to the plasma treatment, the vacuum in the chamber was maintained to 0.2 mtorr. After the pressure became stable, the plasma treatment was carried out by a radio frequency power generator operating at 80 W for 3 minutes. The plasma modification was done using PE-100 Benchtop Plasma System (Plasma Etch, Inc Drive Carson City, USA) in a 12" X 12" X 12" Welded Aluminum Vacuum Chamber. (Scheme 2). The plasma modified LDPE is coded as PLDPE.



Scheme 2: Plasma modification of LDPE

2.1.2.3. Preparation of LDPE foam composites

LDPE was mixed with 8g (i.e. 4phr) of endothermic blowing agent (a mixture of sodium bicarbonate and citric acid) along with the prepared master batch. The mixture was then extruded using a twin screw extruder (Twin Extruder (KETSE 20/40D - Brabender) with the temperature of zone 1,2,3,4 and 5 as 100, 70, 180, 180 and 190°C. The extruded composite in the sheet foam was then analyzed.

The different foam composites prepared and their code is given in Table 1. The explanations for the codes are as follows. G stands for gum compound without fiber, F stands for fiber filled composite, B stands for blowing agent, M stands for maleic anhydride, P stands for plasma modified LDPE and the digits 3 and 5 indicate the amount of fiber in the composite. For example, FMPB5 indicates a fiber filled composite having 5 phr fiber containing maleic anhydride, plasma modified LDPE and blowing agent.

Table 1 :The different foam composites prepared and their code

CODE	Composition
GB	LDPE + Blowing Agent
FB3	LDPE+ Blowing Agent+3phr Fiber
FB5	LDPE+ Blowing Agent+5phr Fiber
GMB	LDPE+ Maleic Anhydride + Blowing Agent
FMB3	LDPE+ Maleic anhydride + Blowing Agent+ 3phr Fiber

FMB5	LDPE+ Maleic anhydride + Blowing Agent+ 5phr Fiber
GMPB	LDPE+ PLDPE+ Maleic anhydride + Blowing Agent
FMPB3	LDPE+ PLDPE+ Maleic anhydride + Blowing Agent+3phr Fiber
FMPB5	LDPE+ PLDPE+ Maleic anhydride + Blowing Agent+5phr Fiber
FPB5	LDPE+ PLDPE+ Blowing Agent+5phr Fiber

2.2. Characterisation techniques

2.2.1. Mechanical properties

The tensile properties of the films were measured using the RSA-G2 (TA Instruments, USA) according to ASTM D638-5 at 25°C and at across head speed of 10 mm/min. Dumbbell-shaped specimens were cut with a dimension of 65 mm x10 mm x3 mm. Five specimens of each sample were tested; the tensile strength, elongation at break and Young's modulus were calculated as an average of five measurements.

2.2.2. Scanning electron microscopy

Nova Nano SEM field emission Scanning electron microscopy with an accelerating voltage of 5.0 kV and Joel Benchtop SEM- JCM 6000 with an accelerating voltage of 10kV was used. The samples were cryocut in liquid nitrogen and the surface was plasma treated to remove the impurities.

2.2.3. Fourier transform infrared spectroscopy (FTIR)

The FT-IR spectra of the films were measured using a Perkin Elmer Spectrum 100. The Fourier-transform infrared (FTIR) spectroscopic analysis of the samples was done from 400 to 4000 cm^{-1} in a Perkin Elmer Spectrum 400 FTIR with an ATR detector at a resolution of 4 cm^{-1} .

2.2.4. Differential scanning calorimetry

DSC analyses were performed on a Perkin Elmer DSC4000 differential scanning calorimeter. The samples were scanned in two cycles at a heating rate of 10°C/min from 40 to 140°C in nitrogen atmosphere.

2.2.5. Thermogravimetric analysis.

Thermal gravimetric analysis (TGA) was done using a Perkin Elmer Pyris 6 TGA in a controlled atmosphere of nitrogen, with scan range from 35° to 800° at a constant heating rate of 10 °C/min in a continuous nitrogen flow.

3. Results and Discussion

3.1 Characterisation of micro-fiber

3.1.1 Scanning electron microscopy

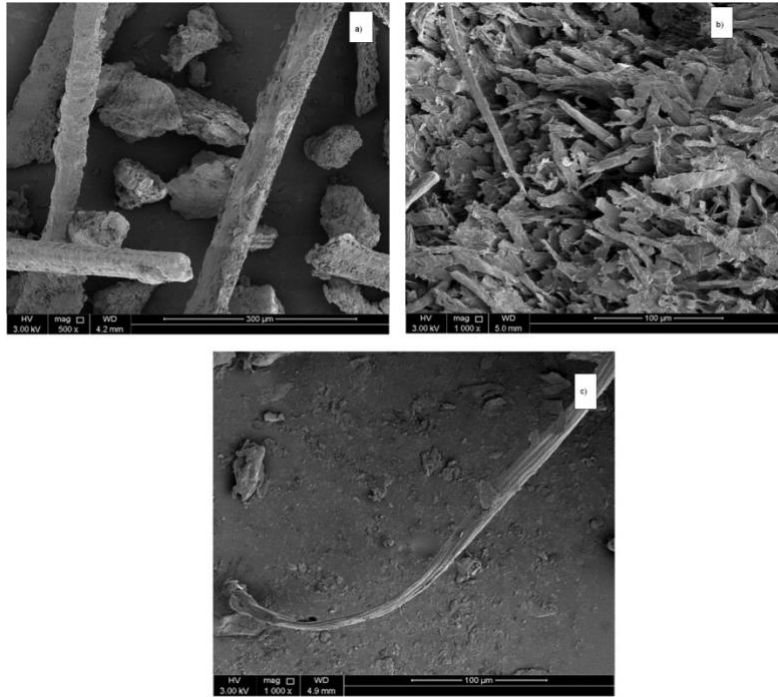


Figure 1(a-b). SEM images of the a) raw fibers, and (b & c) bleached fiber.

The comparison between the treated and untreated fibers shown in Figure 1 (a-b) shows the structural differences affected by bleaching. The arrangement of cellulose fibers within the matrix of lignin is clearly seen in this picture. Figure 1a and b show defibrillation by dissolving the lignin that gets depolymerized. Bleached fiber shows a thickness in the range of 5–10 μm. Each elementary fiber (figure 1 & d) shows a compact structure showing aligned fibers. The analysis shows that microfiber manufacturing using the above-mentioned method was successful. However, the figure shows the presence of some compacted structure which can be due to the presence of residual lignin content.

3.1.2 Fourier transform infrared spectroscopy.

The FTIR spectra of raw fiber and alkali treated fiber are presented in Figure 2, which shows a change in the spectrum. Considerable changes were noticed in alkali treated fibers and are also supported by the SEM image. It is reported that alkali treatment reduces hydrogen bonding by reacting with sodium hydroxide which results in an increase in -OH concentration which is evident from the increased intensity of the peak between 3300 and 3500 cm^{-1} bands as compared to the raw fibers and this corresponds to the stretching of hydroxyl (O-H) groups in cellulose. The peak at 1600 cm^{-1} is related to the absorbed water. The peak appearing around 1508 cm^{-1} is due to the CH₂ stretching from cellulose. The peak around 1236 cm^{-1} is considered due to C=O stretching of acetyl in lignin which disappeared after treatment. The peak at around 1072 cm^{-1} is assigned to C-O deformation in secondary alcohol and aliphatic ether. The peaks around 1515 cm^{-1} and 1240 cm^{-1} seen in untreated fiber disappear after alkalization. The disappearance of the peak around 1515 cm^{-1} indicates that the non-cellulose such as pectin, and fats are removed during the alkalization. The

disappearance of the peak around 1240 cm^{-1} after alkalization indicated that the removal of hemicelluloses[22,23,24]

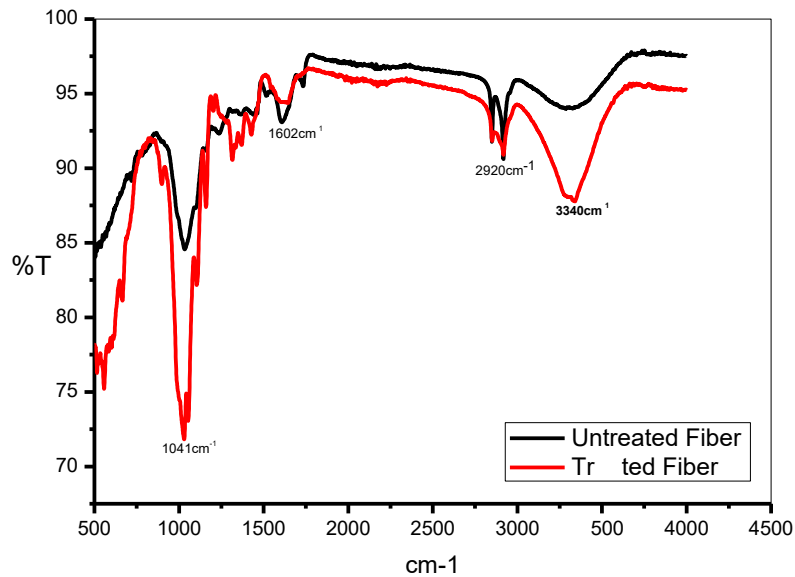


Figure 2. FTIR spectra of raw fiber and alkali treated fiber

3.2 Characterisation of foam composites

3.2.1 Effect of filler loading

3.2.1.1 Morphological Analysis

Figure 3 shows the SEM analysis of foam composites with different fiber loading and compatibilizers. The figure shows that adding 5phr of fiber without any compatibilizer, increases in the number of cells dramatically. It can be observed from Figure 3a and b that on adding fibers, the cell size of the foam decreases. Foamed plastics exhibit reduced apparent densities due to the presence of numerous cells dispersed throughout the mass of the polymer. Foamed samples of all composites fiber show cells with finer average sizes in comparison to those of virgin LDPE.

Figure 3 c, d, e&f shows respectively the SEM micrographs of the 5 phr fiber reinforced foam composites with *PE-g-MA* as the compatibilizers, hybrid compatibilizer of *PE-g-MA* & plasma modified LDPE and with plasma modified LDPE (PLDPE) alone. The micrographs show that for the foam composites the addition of compatibilizer causes a change in the shape of the cells. However, cell density is found to decrease for compatibilized foam composites and the addition of the compatibilizers reinforces the foams which are confirmed by the mechanical property data.

The size distribution of the pores for each sample was found by measuring the area using ImageJ software and is plotted in figure 3g. The comparatively smaller pore size was found for the composite when maleic anhydride alone was used as the compatibilizer. However for all the composite the pore size was found to be reduced compared to the sample without natural fiber. The mechanical properties of foam depend on the cell structure and cell distribution of foam. [25] However, for plasma modification alone, the cell shape seems to be the same although the mechanical properties were improved by plasma modification alone showing an improvement in the interfacial adhesion.

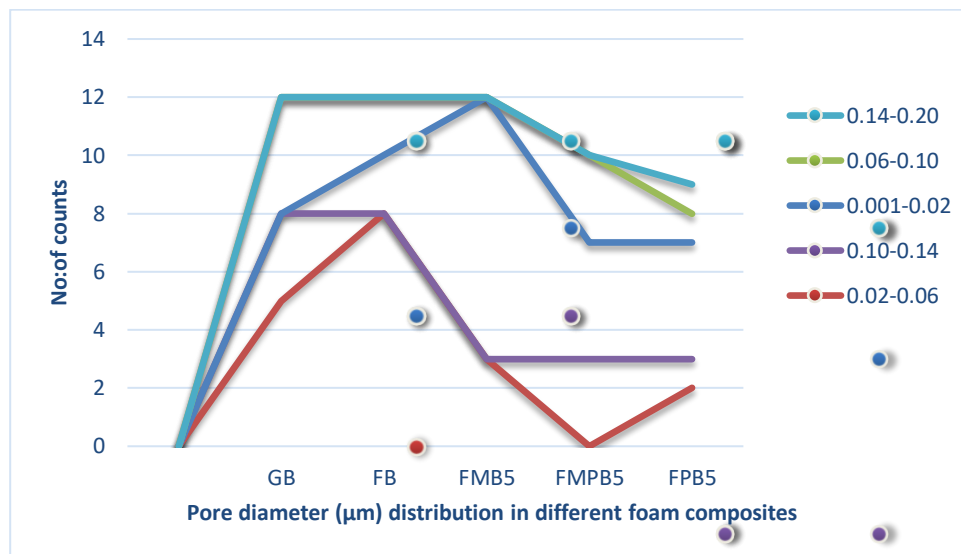
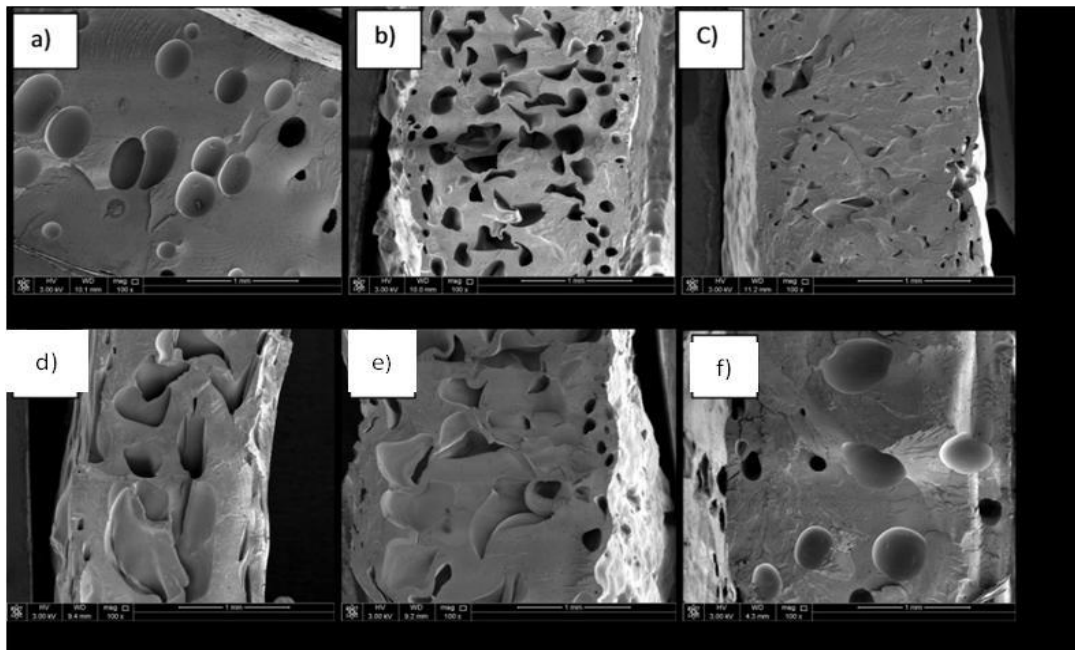


Figure 3.1. SEM images of the foam composites a) GB without any fiber b) FB5 with 5phr micro fiber c) FMB3 with 3phr fiber and *PE-g-MA* as compatibilizer d) FMPB3 with 3phr fiber and with hybrid compatibilizers (*PE-g-MA* and PLDPE) e) FMPB5 with 5phr fiber with hybrid compatibilizers (*PE-g-MA* and PLDPE) f) FPB with 5phr fiber and PLDPE g) The pore size distribution in each composition

3.2.1.3 Mechanical properties

Figure 4A shows the stress-strain behaviour of the foam composites without filler (GB 5) with 5 phr microfiber alone (FB5), and with 5 phr micro fiber with PLDPE (FPB5). It is reported that the mechanical behaviour of polymer foams are dependent on the density, size, and shape of the cells and of the nature of the material,[26] The value shows that for the foam composite with plasma modified LDPE, the tensile strength was improved showing the better adhesion of fiber and polymer. The decrease in a number of cells in the case of plasma modified composite is also a reason for its influence on the tensile strength. The interaction between the fiber and the polymer in the case of *PE-g-MA* incorporated foam composites results in the deformation of cells. Figure 4B shows the stress-strain behaviour of the foam composites with *PE-g-MA* as the compatibilizer without filler (GB 5) with 3 phr microfiber (FMB3), with 5 phr micro fiber (FMB5). Compared to neat foam composite, it can be observed from the stress-strain graph of the 3 and 5 phr fiber (with *PE-g-MA* as the compatibilizer) that fiber incorporation could reinforce the foam composite to a good extent. The value of tensile strength is almost doubled with a higher toughness for the 5phr sample. Figure 4C shows the stress-strain behaviour of the foam composites with both *PE-g-MA* and plasma modified LDPE as the compatibilizer without filler (GMPB 5) with 3 phr microfiber (FMPB3), with 5 phr micro fiber (FMPB5). For both *PE-g-MA* and plasma modified LDPE incorporated foam composites, it is observed that the presence of both *PE-g-MA* and plasma modified LDPE have effected better reinforcement at 3 phr fiber loading. However, the toughness of both composites was lower than neat foam composites. This is due to the availability of interaction sites which is explained later in each paper. An addition of more compatibilizer can affect in more property and this can be a scope for future studies which can be done by varying the compatibilizer concentration. The increase in a number of cells as observed from the morphological data (SEM) for the hybrid compatibilizer incorporated foam composites agrees with this. The incorporation of 5phr fiber shows better toughness. The increase of the maximum strength and the modulus due to the addition of compatibilizers

was due to an improvement in the mechanical adhesion that is promoted by the generation of greater surface area and roughness on the fiber as a result of the plasma treatment, the increase in the compatibility between the fiber and the matrix which decreased the fibres' polarity, the formation of more bonds with the hydroxyl groups (OH) of the fiber and the organofunctional group with the polymer. An explanation of this is given later in the paper

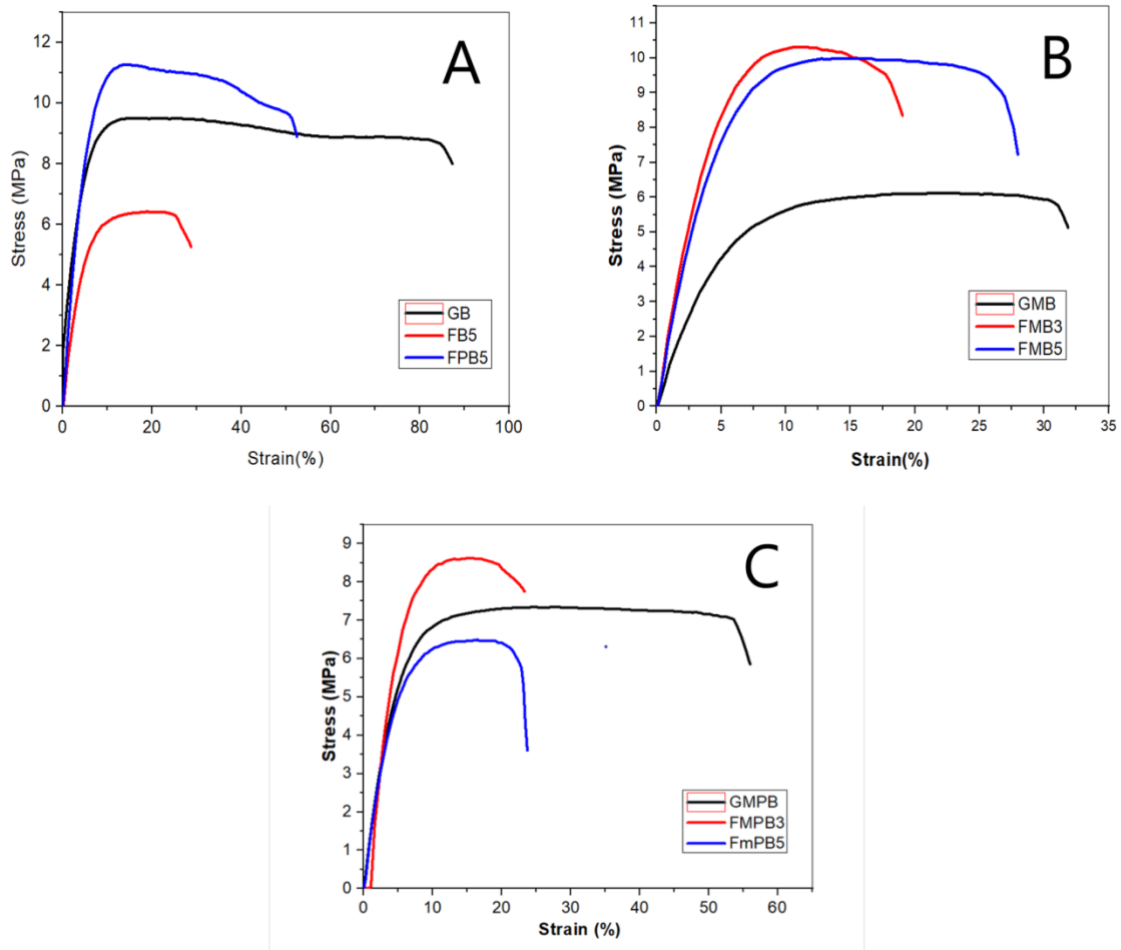


Figure 4. A) Stress-strain graph of the foam composites with different fiber loading A) without any compatibilizer B) with *PE-g-MA* compatibilizer C) with hybrid compatibilizers (*PE-g-MA* and PLDPE)

3.2.1.2 Differential scanning calorimetry (DSC)

The DSC thermogram of the foam composite with various loading is given in figure 5. LDPE contains both crystalline and amorphous regions. The crystalline region consists of spherulites where the polymeric chains form an organized structure.²⁷ The short branches in LDPE lead to the formation of amorphous regions. From the DSC analyses during heating it is possible to determine the melting point, level of crystallinity and the presence of different phases in semi-crystalline polymers. The figure presents exothermal curves of foam

composites at different cooling rates. It could be observed that incorporation of fibers makes crystallization peak temperature (T_c) move slightly to higher temperature compared to neat foam composites, indicating some nucleation effect of fibers. The increase in crystallization temperature associated with the addition of micro and nano fillers in semi-crystalline polymers has been extensively reported (ref). The change confirms that fibers affect the phase formation in LDPE crystallization and shows that it affects the crystallization behaviour too. The level of crystallinity of PE was enhanced with the incorporation of fibers. Moreover the nucleating effect is stronger for compatibilized system where particles are better dispersed than for uncompatibilized system where the dispersion of the fibers was poorer.

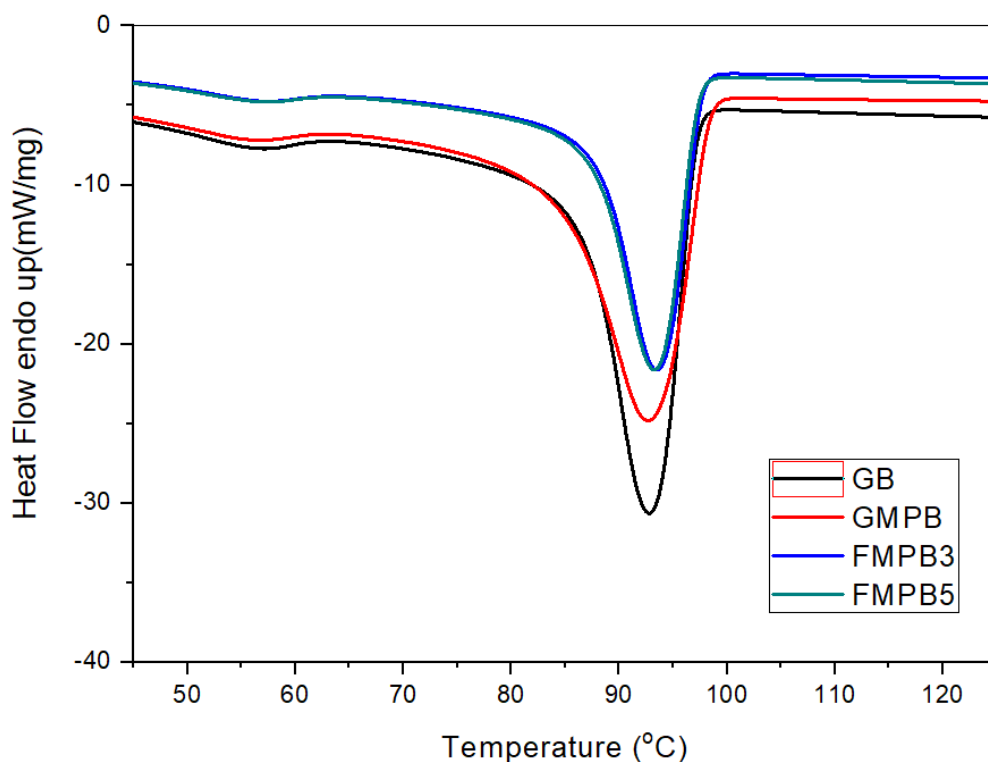


Figure 5. DSC thermogram of images of the foam composites -GB without fiber, GMPB foam composite without fiber, MPB3 with 3phr fiber and hybrid compatibilizers(*PE-g*-MA and PLDPE), FMPB5 with 5phr fiber and hybrid compatibilizers(*PE-g*-MA and PLDPE)

3.2.1.4 Thermogravimetric analysis (TGA)

Thermograms of different foam composites are shown in figure 6. It is evident from the figure that the thermal decomposition of plasma modified foam composite with 5 phr fiber loading is comparatively higher showing more number of fiber polymer interactions and good adhesion between fiber and the polymer when PLDPE is added. This increased thermal stability indicates that the incorporation of the fibers into the material introduces more

number of nucleation points that increase the crystallinity of the polymer, and, which then results in improved thermal properties. However, the presence of fibers in the foam composite with *PE-g-MA* is not showing any increase thermal stability. This difference can be described to be due to the nature of the interaction between fiber and LDPE on varying the nature of compatibilizer which will be discussed in the next section.

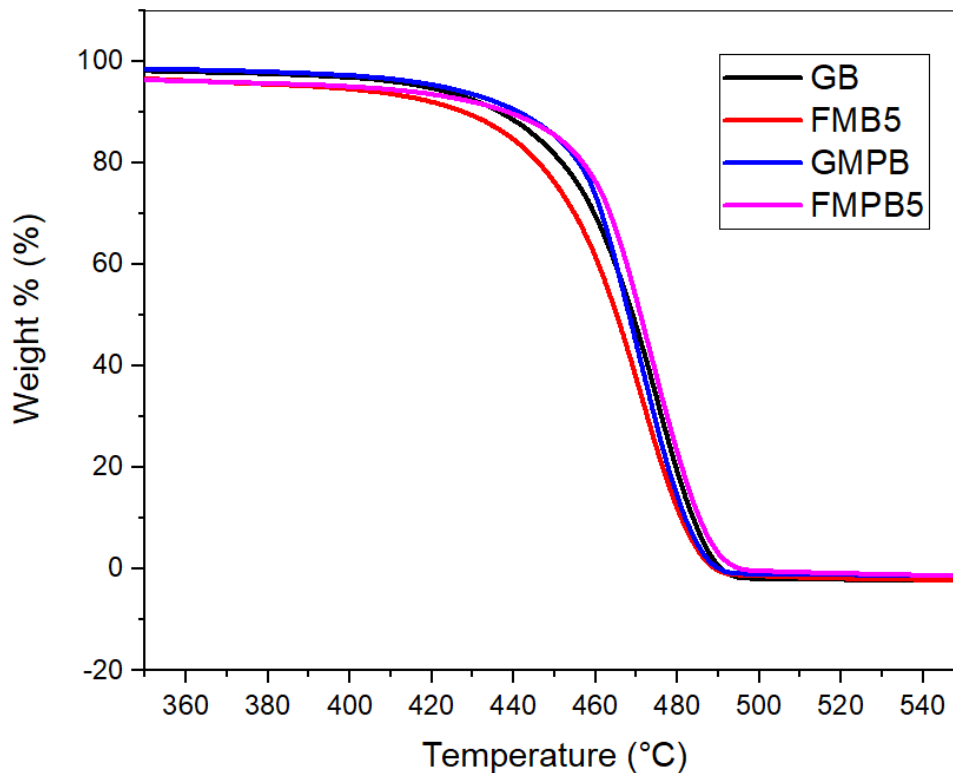


Figure 6. TGA curve of the foam composites

Pl correct the Y axis of the figure 6, 2 times percent appeared.

3.2.2 Effect of plasma modifications and its hybrid effect

The morphological, thermal mechanical and thermo mechanical analysis of the foam composites with and without fibers and different compatibilization methods show that compared to other foam composites, the plasma-treated LDPE based foam composite could improve all the properties. The morphological interpretation of the SEM images showed more cells in the case of foam composites with no compatibilizer. However there was a noticeable change in the cell shape upon adding *PE-g-MA*. Although the cell density values were comparatively decreased for the foam composite with PLDPE, the composite showed excellent reinforcement with appreciable cell density. The difference in the shape of the cells compared with *PE-g-MA* can be due to the difference in the nature of the interaction. As a

result of the superior compatibilization process arising from the plasma surface treatment, the LDPE matrix is well bonded to the fiber surface. This in fact caused the deformation of the cells. The thermal properties showed comparatively more thermal stability for the foam composite with PLDPE (Fig 8a). The mechanical property (Fig 8b) also could support superior reinforcement when PLDPE was used. Both the tensile strength and toughness were improved. This improvement in PLDPE incorporated foam composites is supported by the well-known fact that the plasma treatment of the polymer surface results in the creation of new reactive sites on the polymer surface and can produce a variety of atomic functional groups, like carbonyl, carboxyl, and ester groups etc at the surface. Usually plasma treatment results in two simultaneous processes, one which increases the surface roughness by the process of etching of the polymer surface and other the formation of functional groups at the polymer surface by the interaction between the active species from the plasma and the surface atom. Such a modification for polymer is favourable for subsequent grafting on account of increased surface area. Thus the presence of both the surface roughness and that of the polar group on the LDPE must have caused superior interaction with the natural fiber and this must have thereby increased the adhesion between the two. However, on comparing the maleic anhydride grafted PE and the PLDPE foam composites the improvement is quite higher for PLDPE foam composites and this can be verified by the fact that, while for maleic anhydride grafted PE, the two carbonyl groups in each of it causes an interaction with the hydroxyl group of cellulose as represented (scheme 1) in the case of PLDPE foam composites. In other words, more number of polar group on the surface of the LDPE causes more sites for interaction and thereby resulting in an improvement in the overall properties. In the case of hybrid compatibilizer, the number of sites for the interaction is reduced because of the additional possibility of interaction between maleic anhydride and PLDPE [28,29] as represented in schematic 1, which causes a reduction in the number of interaction sites with the natural fiber.

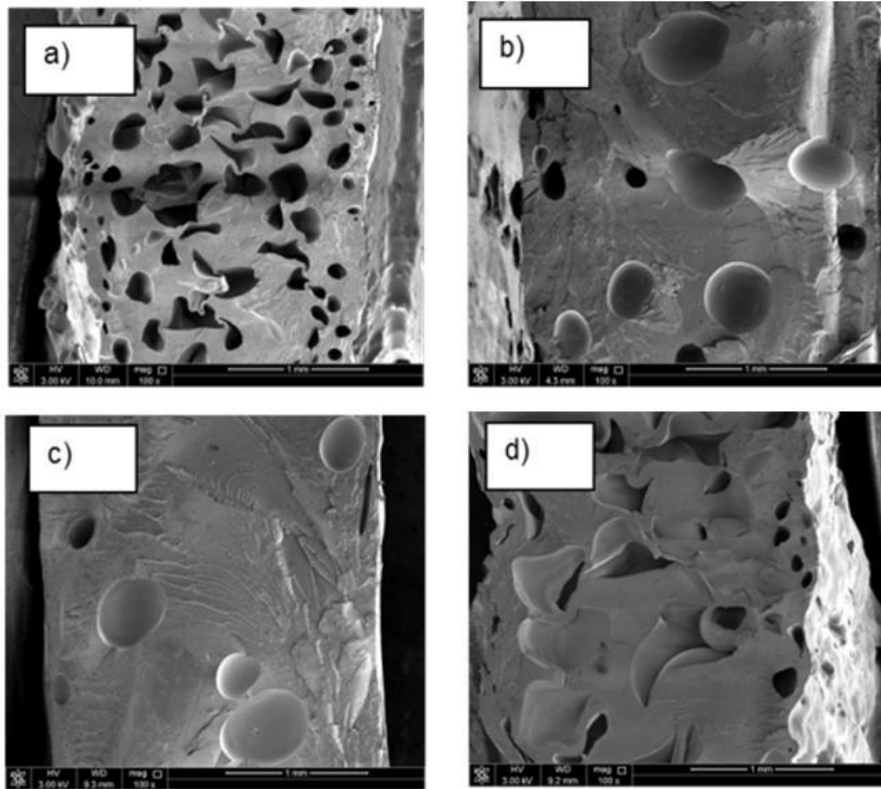


Figure 7. SEM images of 5phr fiber a) FB5-without any comp tibilizer – b) FMB5-with 5phr fiber and maleic anhydride as compatibilizer c) FPB5 with 5 phr micro fiber and plasma modified LDPE and d) FMPB5 with 5 phr micro fiber and hybrid compatibilizers ($PE-g-MA$ and PLDPE)

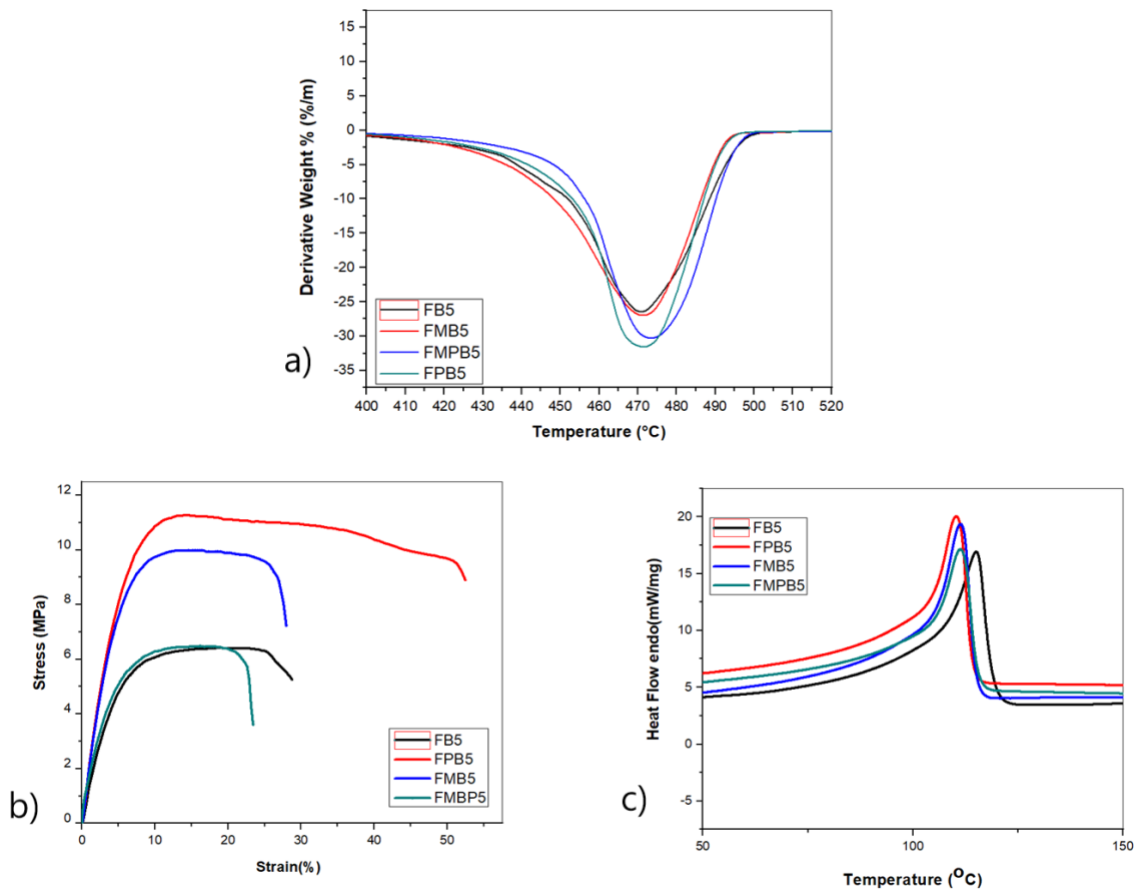
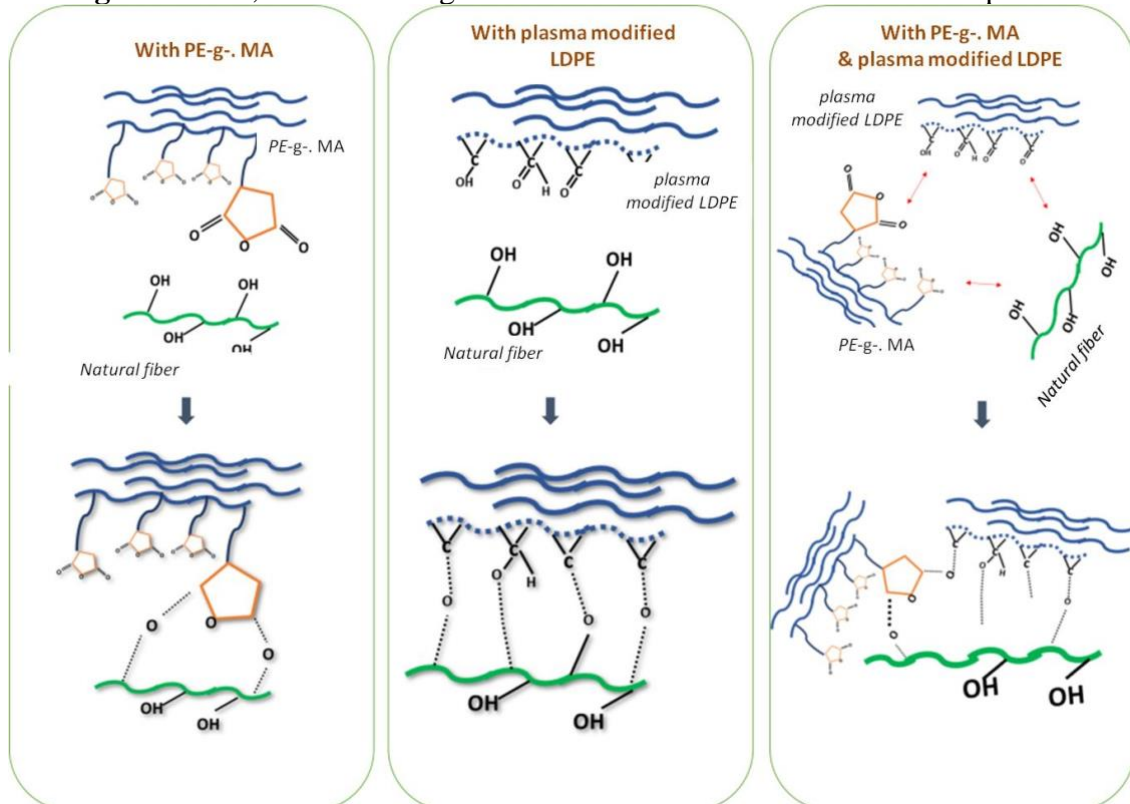


Figure 8.DSC, Tensile strength and TGA data of LDPE foam biocomposites



Scheme 1: Possible interaction between the compatibilizers and natural fiber

4. Conclusion

From the overview of the results, it was found that both *PE-g-MA* and plasma modified LDPE (PLDPE) could impart modification and could reinforce the foam composites. In fact, three different types of interfaces have been created using *PE-g-MA*, PLDPE and a combination of *PE-g-MA*, PLDPE. Among them, PLDPE gave the best results. Morphology of the foam composites revealed that the difference in fiber loading and compatibilizers could show a relevant modification in the foam morphology and microstructure. The mechanical property data and thermal analysis could support the better adhesion in the case of PLDPE incorporated foam composite and the study could bring about the idea that the incorporation of PLDPE as the compatibilizer is an environment-friendly method and could reinforce the natural fiber-LDPE foam composite. The study gives scope for improvement of other properties like cell density, cell size etc by varying the other parameters. Studies are in progress to scale up the process to commercial scale.

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