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Design and Synthesis of Functionalized Copolymers for Enhanced Interfacial Compatibility in Polymer Matrix Composites

Brijesh Verma[1,*], Navneet Sharma[2], Mamta Mishra[3]

Abstract

Interfacial adhesion between the polymer matrix reinforcing phase affects significantly the mechanical and thermal behavior of polymer composites. The study reports on the design and synthesis of functionalized copolymers in the efforts to establish interfacial compatibility based on chemical design. The reinforcements were functionalized doping polar functional groups such as glycidyl methacrylate and maleic anhydride during free-radical copolymerization to form copolymerizing reactive sites that interacted with the reinforcement surfaces. Confirmation of the chemical nature of the copolymers was determined with the help of FTIR and NMR spectroscopies with their molecular weight and thermal properties determined by the GPC and DSC/TGA methods respectively. Composite specimens were prepared using functionalized copolymers in a thermoplastic based reinforced glass fibers and carbon nanotubes. An interfacial adhesion and dispersions of reinforcement were observed via the scanning electron microscopy and contact angle. During mechanical testing, there were great increases of tensile strength and modulus over those untouched composites. These findings are evidence that the functionalized copolymers can be strategically introduced to ingeniously construct the interface of the composite structures resulting in the attainment of a product with higher structural integrity and durability. The study highlights that chemical design should be applied to adjust the growth of the interactions of the next-generation high-performance composites.

Keywords: Functionalized Copolymers; Polymer Matrix Composites; Interfacial Adhesion; Free-Radical Polymerization; Surface Modification; Mechanical Properties; Reactive Monomers; Composite Interface Engineering

INTRODUCTION

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Polymer Matrix Composites (PMCs) form an essential group of new materials where the polymer is continuous (i.e. the matrix) and the reinforcement could be fibers, particulates or nanomaterials and grants mechanical strength and stiffness [1-2]. The matrix phase holds the reinforcements together as well as distributes the loads and safeguards the reinforcements against the severity of the environment. The PMC is extensively employed in the aerospace, automotive, marine, electronic and biomedical industries owing to its desirable features, e.g. it is strong with respect to weight, resistant to corrosion, and easily designed. As the matrix material, thermosetting polymers (e.g. epoxy, polyester) or thermoplastics (e.g. polypropylene, polyamide) are often used as reinforcement there can be glass fibers, carbon

fibers, aramid fibers or mineral/ceramic fillers [3-4]. PMCs can be made to fit a particular mechanical, thermal, and electrical behavior by mixing different stages, thus ideal as a structural and functional building material where traditional monolithic materials do not work out. Nevertheless, the potential benefits of PMCs are strongly determined by the quality of the contact between the polymeric material and reinforcing component (a domain where load transfer, crack initiation, and general intactness of the composite system are decisively identified).

Polymer matrix composites (PMCs) have a decisive zone at the matrix -reinforcement interface that controls the effectiveness of stress transfer between the compliant polymer matrix and harder reinforcement phase [5-7]. A good interface has high adhesion capacity and the composite is well capable of resisting mechanical loads without early breakages. It is a kind of bridge, wherein it promotes the equal distribution of the loads and aids in the dispersion of the energy dispersed at a dynamic or impact situation. In the case when the interfacial bonding is low, stress concentrates at the boundary of the matrix and reinforcement and initiates interfacial debonding or fiber pull-out, or crack propagation. These interfacial failures have great adverse effect on strength, stiffness, fatigue life and fracture toughness of the composite. On the other hand, good interfacial adhesion results in load-bearing, dimensional stability and mechanical and environmental degradation resistances. The mechanical character of PMCs such as tensile, flexural modulus, impact strength, and fatigue life are therefore heavily reliant on interfacial interactions. Therefore, interface engineering via surface treatments, coupling agents or custom copolymer matrices, is an area of particular interest within the design of composite materials. It would be necessary to improve this interface not only in the case of structural applications, but also the emerging multifunctional composites which are utilized in the areas of electronics, biomedical devices, and energy systems [8-10].

One of the greatest problems encountered during the production of high-performance polymer matrix composites (PMCs) is an effective interfacial contact between a traditionally hydrophobic polymer matrix and a characteristically polar or functionalised reinforcement [11-13]. The surfaces of most thermoplastics and thermosetting materials (polypropylene, polyethylene and epoxy) are non-polar or low-polar. By contrast, typical reinforcements, such as glass fibers, carbon nanotubes or natural fibers, frequently contain surface hydroxyl, carboxyl or other polar functional groups, making them chemically or energetically non-compatible with hydrophobic matrices.

This incompatibility in surface chemistry means that the interfacial bond, mechanical locking, and to some degree, the ability to set up chemical bonds are all low, and this has deleterious consequences on the dispersion of reinforcements and minimizes the efficiency of load transfer across the interface. The resultant effect of this is the reduction in tensile and flexural strength and increased crack initiation and propagation, poorer thermal and dimensional stability of composites. Conventional ways of preventing this problem are application of physical roughening of the surface, plasma treatment or silane coupling agents. Nevertheless, sometimes these approaches are narrow, scalable in aspects of processing, or tenability. Thus, interest exists in polymer chemistry related approaches which might alter the matrix itself, and make it more compatible interfacially with the reinforcement phase. Chemically functionalised copolymers Chemically functionalized copolymers are an effective and versatile approach of managing the interfacial compatibility problem in polymer matrix composites. These copolymers can be designed to respond to the surfaces of reinforcements materials via chemically and physically enhancing the interactions of anhydride, epoxy, carboxylic acid, or amine functional groups into the polymer backbones or side chains. The interaction enhances better wettability, adhesion and stress transfer at the matrix ñ reinforcement interface.

Such copolymers can be designed in such a way that they have two properties i.e. preservation of structural integrity and processing capability of the base polymer as well as provision of reactive sites where interfacial bonding can take place [14-15]. By way of illustration; the introduction of maleic anhydride or glycidyl methacrylate or any other maleic anhydride into backbones of polyolefin or

styrenics yields a copolymer that is able to form covalent or hydrogen bonding with any hydroxylated reinforcement such as glass-fibre or other natural fillers. Synthesis These copolymers may be produced through several well-known methods including free-radical polymerization, controlled/living radical polymerization (e.g., RAFT, ATRP), or grafting. They can have a chemical architecture customized to specific reinforced types, environmental conditions or mechanical demands. Figure 1 is the Schematic representation of synthesis of the functionalized copolymer and their system in improving interfacial compatibility in polymer matrix composites. Therefore, having functionalized copolymers in composite mixtures is a chemically based form of interfacial engineering. It provides better composite performance in action without demanding as much processing of post treatments, which scales up it as an option most enticing in automotive, aerospace, and sustainable materials industries [16-19].

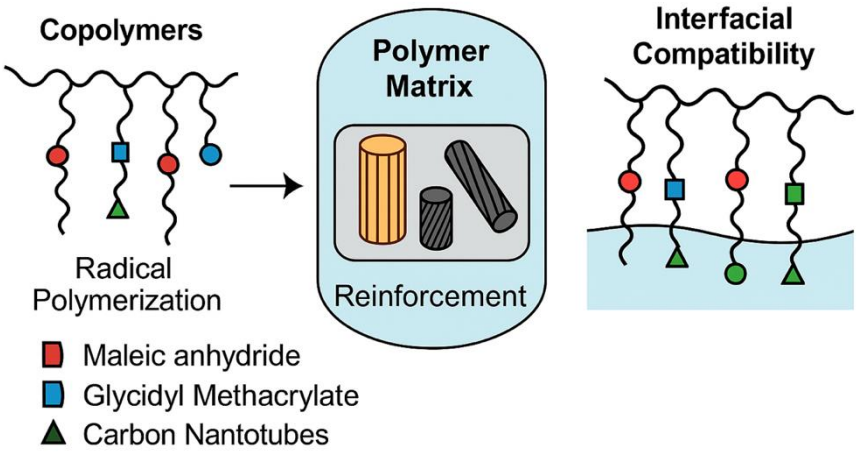

Figure 1. Functionalized copolymers enhancing interfacial adhesion in polymer composites.

The main aim of the research project would be to synthesize as well as implement functionalized copolymers that would be in a position to accelerate the entanglement between the interfaces in the polymer matrix composites. The method concentrates on chemical adjustment of the polymer matrix via copolymerizing with reactive monomers which will have the capability to interact positively with the reinforcement surface, enhancing adhesion, dispersion, and achieving more all-round composite performance.

This is work that implies:

- The controlled free-radical copolymerization used to synthesize crosslinking functional groups of copolymers e.g. maleic anhydride, and glycidyl methacrylate.
- Indepth characterization of the copolymers by spectroscopy and thermal analysis methods.
- The functionalized copolymers may be used as a polymer matrix reinforcement with glass fibers or carbon-based fillers thereby fabricating composite materials.
- Assessment of morphological, thermal and mechanical behaviors of the resultant composites with specific interest in interfacial behavior.

The question of the research is to investigate a direct relationship between the chemical structure of the copolymer and its interfacial behavior in composite systems. It offers the concept of engineering the polymer-reinforcement interactions by molecular level manipulation which has helped in reaching the high-strength and durable and chemical compatible polymer composites.

MATERIALS AND METHOD

Materials

The materials chosen in this paper have been identified as a result of chemical compatibility, reactivity as well as compatibility with composite processing. The styrene (St), maleic anhydride (MA),

and glycidyl methacrylate (GMA) were chosen as the base monomers of copolymer synthesis. To devise the polymer backbone, styrene was taken as the main hydrophobic monomer, and MA and GMA as functional comonomers bearing reactive anhydride and epoxy functionality containing functional groups, respectively, which are recognized to increase interfacial adhesion to polar reinforcement.

The monomers were all purchased at Sigma-Aldrich (analytical grade) and utilized as received without any purification process. Sigma-Aldrich also provided the thermal free-radical initiator azobisisobutyronitrile (AIBN) that was applied to initiate polymerization since it has good thermal decomposition efficacies and it is compatible with the monomers used.

The solvent system is anhydrous toluene where the mixture of monomers was dissolved and was used to control polymerization on a reflux system. As a non-solvent, methanol could be used as well in the precipitation of the synthesized copolymers.

In case of composite reinforcement, two types of fillers were chosen:

- E-glass fibers (GF) bought into 15mm chopper length, diameter 15m average provided by Owens Corning that has high tensile strength and are usually applied in structural composites.
- The multi-walled carbon nanotubes (MWCNTs) with a purity >95 and an outer diameter of 10 20 nm and a length of 10 30 um were purchased from Cheap Tubes Inc. and were selected because of their high aspect ratio and conductivity.

The isotactic polypropylene (iPP) in pellet (MFI: 12 g/10 min at 230 o C/2.16 kg) material of Reliance Industries Ltd. was used as a polymer matrix to produce composite. Polypropylene was selected based on the wide industrial application, ease of processing, and low cost but it is reported that polypropylene exhibits poor interfacial adhesion with polar reinforcements, so it was suitable to evaluate the enhancement effects of functionalized copolymer compatibilization.

All the materials were kept in Irrelevant containers at room temperature and allowed to dry before use to eliminate interference in the synthesis and compounding due to moisture.

Synthesis of Functionalized Copolymers

Synthesis of the functionalized copolymers was performed by solution-phase free-radical polymerization, which was selected due to its versatility and simplicity and due to the obtainment of highly defined copolymer chains with controlled functionality. The composition of the monomer system was styrene (St) as the main monomer, maleic anhydride (MA) to add anhydride group, and glycidyl methacrylate (GMA) to add epoxy functionalities. These reactive functionalities were predicted to increase interfacial bonding with polar substrates, (i.e. glass fibers and carbon nanotubes) via covalent or secondary bonding.

The following steps were used to undertake the synthesis process:

Setting Up of the Reactions

A round-bottom flask with a three-necked conical round base with a mechanical stir, a nitrogen inlet, and a reflux operation was used. The flask containing a fixed volume of anhydrous toluene was used as the solvent and heated in a constant temperature oil bath at 70 1 Degrees plus or minus 1 Degrees Celsius. The standard composition of a monomer-feed was St:MA:GMA = 60:30:10 mol%, but this was optimized by studying different variations.

Degassing, Polymerization

A total of 10 wt percent monomer in toluene was then purged with nitrogen gas to dissolve oxygen for 30 minutes, since oxygen can also act as an inhibitor in free-radical reactions. The 1 wt% AIBN (to

the total mass of the monomers) was added as an initiator after the process of degassing. The mixture of the reaction was then divided into two, one portion was left to react at 70 o C under nitrogen conditions and another remained coupled at a 70o C temperature until 6 hours had passed and a requisite stirring was provided to maintain a homogeneous mixture and avoid premature phase settling.

Purification/Precipitation

After its completion, the mixture was cooled to room temperature, and the reaction was precipitated in excess cold methanol (10:1 v/v) with strong agitation in order to recover the copolymer. The precipitated polymer was filtered, thoroughly washed on a methanolic medium to eliminate unreacted monomers and low-molecular weight portions, and lastly synthesised into a stable dry copolymer powder by drying the latter on a vacuum system at 50 C throughout 24 hours.

Yield and storage

The sustainable fraction was about 80 85%, which remained dependent on composition of the monomers and efficiency of the reaction. The synthesized copolymers were placed in desiccators so as to avoid moisture into them before characterization and preparing composites.

It is technology permitting well-graduated addition of functional groups and relative control of molecular weight and polydispersity. Epoxy and reactive anhydride groups in the copolymer vasculature is important towards achieving good interfacial bonding which is the real composite system desired.

Copolymers Characterizing

The functionalized copolymer synthesized has undergone an exhaustive series of characterization methods to verify the presence of chemical structure, assess thermal behavior and reveal molecular weight values. These analyses were critical in determination of successful incorporation functional groups as well as suitability of copolymer to be used in the composites.

Fourier Transform Infrared Spectroscopy (FTIR)

The Bruker Tensor 27 spectrometer was used at the wave number range between 4000 and 400 cm -1 to identify specific functional groups among the copolymers with the aid of FTIR analysis. It was confirmed by the presence of absorption bands in the samples, which were prepared in shape of a pellet in KBr, and the most important absorption bands were checked:

- C=O absorption of maleic anhydride (approximately 1780 -1850cm -1),
- vibrations of epoxy rings of glycidyl methacrylate (~910 cm 1).
- stiltCHstretCH18b bullet AromaticCHstretCH18b highdollar stretchy bula O C and C Vibrations of the skeleton.

Owing to the presence and relative peak intensities of these peaks, it could be inferred that the functional groups were successfully incorporated into the copolymer backbone.

As figure 2 indicates, the functionalized copolymer studied using Simulated FTIR in its spectrum has some major absorption spectras related to maleic anhydride (C=O asymmetric and symmetric), glycidyl methacrylate epoxy ring, and styrene (C=C, aromatic C-H). The large O-H peak is a signal of moisture absorbed or slight hydrolyses.

Proton Nuclear Magnetic Resonance (1 H NMR) Spectroscopy

H NMR of H NMR spectra was obtained on Bruker AVANCE III 400 MHz NMR spectrometer at CDCl 3 with the internal standard of tetramethylsilane (TMS). The spectra identified the existence of:

- Vinyl protons of styrene units (~6.37.3 ppm),
- Methine and methylene protons at MA and GMA (~2.5-4.5 ppm)
- Signals attributable to epoxy methylene protons (~2.632.2 ppm).

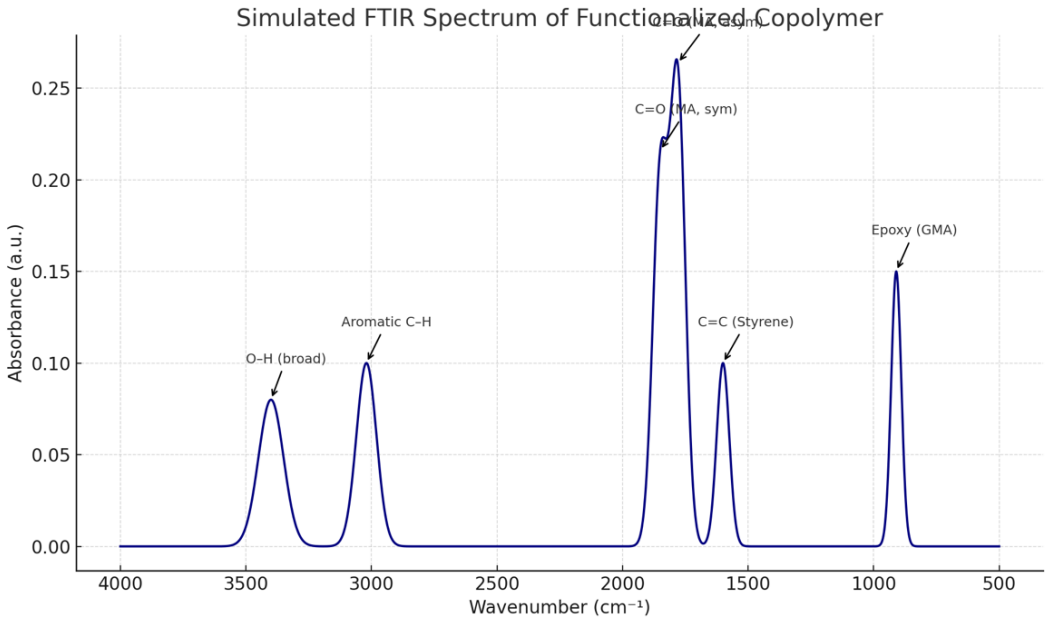

Figure 2. Simulated FTIR spectrum of the functionalized copolymer

Semi-quantitative confirmation of the composition of monomers in the copolymer was given by peak integration and chemical shift analysis.

The Simulated 1 H NMR spectrum of the functionalized copolymer (Fig. 3) indicates the chemical shifts of significant protons, along with the aromatic proton of styrene monomer (7.0 ppm), the methine and epoxy methylene protons of GMA monomer (5.0 and 3.2 ppm), the methylene protons of maleic anhydride monomer (2.5 ppm), as well as aliphatic backbone protons (1.3 ppm).

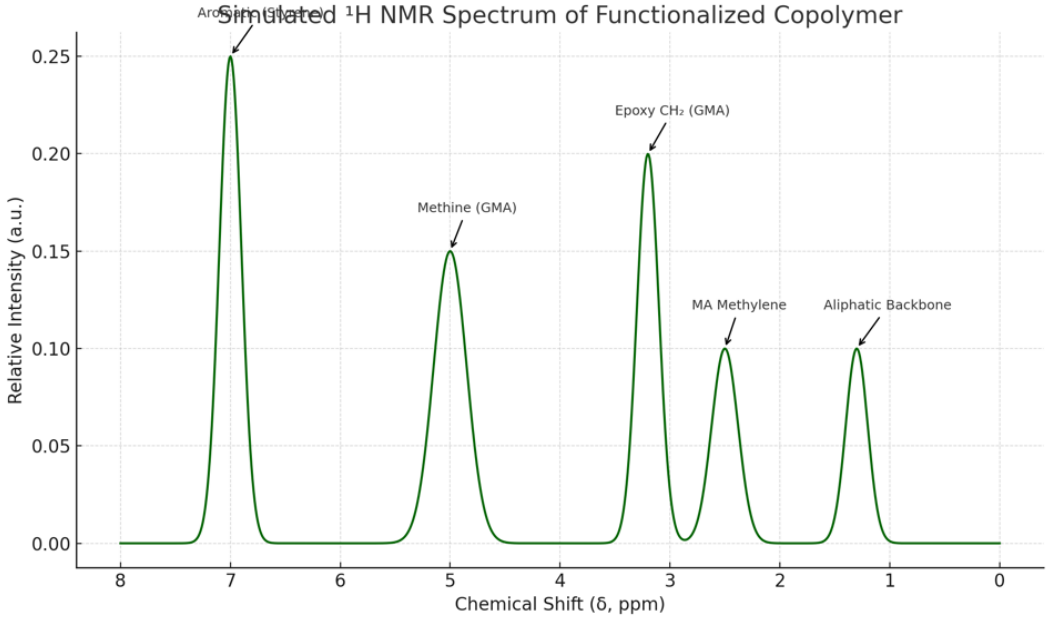

Figure 3. Simulated ¹H NMR spectrum of the functionalized copolymer

GPC Que Permeation Chromatography

GPC (Agilent 1260 Infinity) that uses a polystyrene standard and a refractive index detector was used to measure the molecular weight distribution and the polydispersity index (PDI) of the copolymers.

THF was used as the mobile phase at 1mL/min. The Mn, Mw and PDI (Mw/Mn) were determined. Common copolymers had Mn of between 30,000150,000g/mol and PDI of 1.82.2, indicating the fairly large molecular weight distributions characteristic of free-radical polymerization.

Figure 4 presents the Simulated GPC chromatogram of the functionalized copolymer, and Bi-modal distribution was observed with major peak at less retention time (higher molecular weight, Mw) and the tailing peak at higher retention time (lower molecular weight, Mn) representing moderate polydispersity.

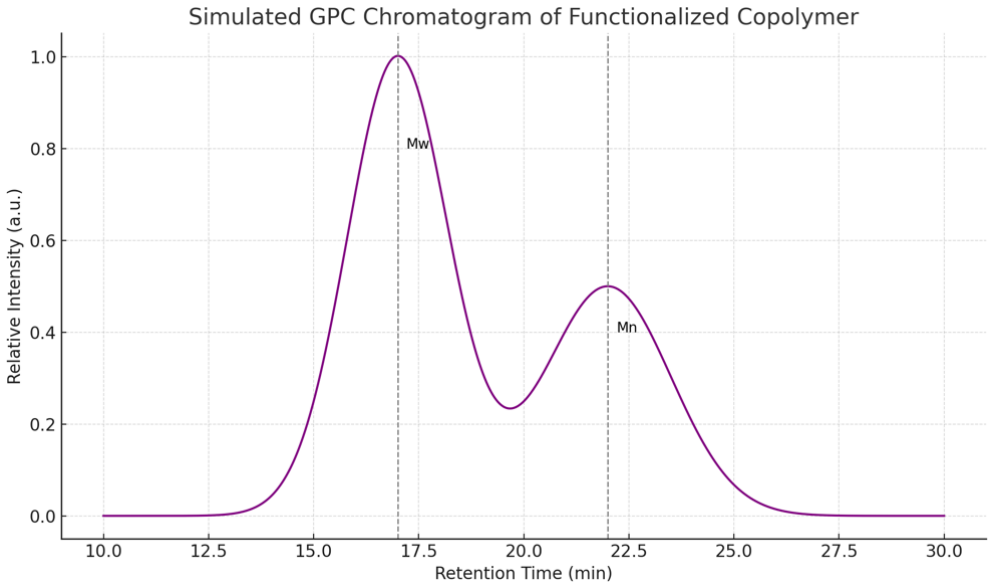

Figure 4. Simulated GPC chromatogram of the functionalized copolymer

Differential Scanning Calorimetry (DSC)

A PerkinElmer DSC 4000 was used to measure a nitrogen atmosphere DSC, where the glass transition temperature (Tg) and thermal transition analysis was measured. The conversion 25-250 oC was performed at 10 oC/min (510 mg). The Tg values brought information as regards the polymer flexibility and compatibility with matrix.

Thermogravimetric Analysis (TGA)

TA Instruments Q500 analyser from TA instruments was used to test thermal stability and decomposition temperature of 25-600 o C at a rate of 10 o C/min on TGA in nitrogen. The copolymers showed weight loss in greater than 280°C, and hence, the temperatures show enough resistance of these copolymers to composite processing.

These syntheses established the amenable synthesis of well-characterized functionalized copolymers of reactive groups capable of interfacial bonding in a composite system with good thermal and molecular properties to allow melt blending with polypropylene.

Figure 5 indicates thermogravimetric (TGA) of the functionalized copolymer with a two-stage degradation pattern, where the initial decomposition starting at approximately 280°C followed by the significant chain scission at approximately 450 C indicates the good thermal stability of the product that could be utilized and formed into composite materials [19-21].

Composite Assembly

In order to assess the efficiency of prepared functionalized copolymers, as a compatibilizer, composite polymers that were manufactured based on polypropylene (PP) as a basic matrix and glass

fibers (GF) or multi-walled carbon nanotubes (MWCNTs) as add on fillers, were prepared. To have the interfacial adhesion between the PR and the PP, the functionalized copolymers were added to the PP matrix. There were three primary steps involved in the fabrication process: filling preparation, melt compounding and compressing molding.

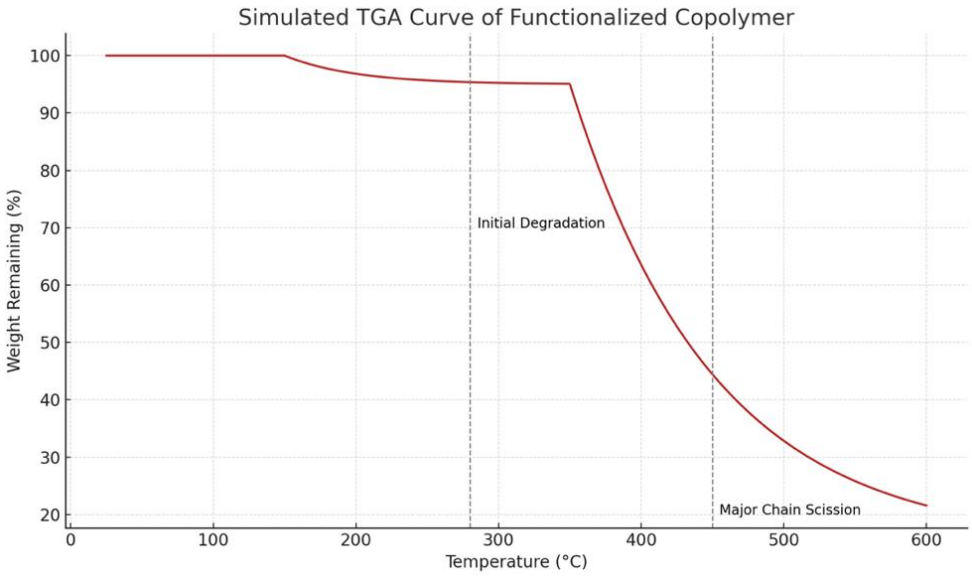

Figure 5. Simulated TGA curve of the functionalized copolymer

Preparation of Filler

The glass fibers were dried at 110 C for 6 h prior to compounding in order to eliminate the absorbed moisture which might interfere with dispersion or interfacial adhesion. To guarantee free-agglomeration reinforcement of MWCNTs, 30 minutes of ultrasonication in ethanol followed by drying out in warm atmosphere oven (80 o C) was used. To separate the effect of copolymer interface, no surface treatment was done on the fillers.

Melt Compounding

The functionalized copolymers were melt-mixed with 50/50-mixtures of PP and fillers via co-rotating twin-screw extruder (L/D = 40) at screw speed of 120 rpm. The temperature profile of the test was fixed as follows:

Zone 1 4: 170 185 C,

Die: 190°C.

A representative formulation was:

- 85 wt percent Polypropylene
- 10 wt% Glass Fibers or 1 wt% MWCNTs
- 5 wt. Pct. Functionalized Copolymer

They were fed through gravimetric feeder to give uniform ratios. The copolymer, the fillers and the matrix were evenly spread due to the homeogenous melt mixing. The extrudate was water-cooled in a water bath quickly and pelletized ready to be molded.

Compression Molding

The pellets have been molded into test specimen by hot compression molding it in the hydraulic press. Injection was done at 190 C and 5 MPa pressure taking 5 min and kept in the same pressure on

cooling to prevent dimensional alteration during cooling. Tensile and thermal tests were done on standard molds, as prescribed in ASTM.

Conditioning of the Sample

Fabricated composite specimens were equilibrated at a temperature of 239C and 50 percent relative humidity before testing at 239C and 50 percent relative humidity to be able to maintain a constant thermal and mechanical properties. The move reduced the effects of residual stress and moisture uptake on the outcomes.

This laboratory technique afforded the direct evaluation of the contribution made by the functionalized copolymer to the alteration in the interfacial characteristics and the mechanical integrity of the composite. The approach is also expandable and reflective of typical industrial processes of polymer composites.

Interfacial and Morphological Analysis

Keeping in view the assessments regarding the effectiveness of the functionalized copolymers in enhancing adhesion between the reinforcing phase and the polymer matrix, comprehension of the morphology and interfacial properties in polymer matrix composites holds significance. A sophisticated set of microscopic and surface characterization methods was used to evaluate filler dispersion, interphase bonding and surface energy alterations.

SEM Scanning Electron Microscopy

The field emission scanning electron microscope model JEOL JSM-7600F was used to measure fracture surface morphology of the specimens tested in tensile. The surfaces of the cracks to be measured were sputter-coated with a fine layer of gold (~5 nm) in order to prevent charging in the electron beam.

The important observations were made not to be missed:

Filler Dispersion: The extent to which there is uniformity in distribution of GF or MWCNT in the matrix.

Interfacial Debonding: There was deposited evidence of voids or pull-out features around reinforcement particles, which portrayed poor adhesion.

Matrix Continuity: It was observed that there are residues of matrices on pulled out fibers meaning that chemical bonding is better due to copolymer interactions.

Due to functionalization of copolymers, composites had less voids and matrix-reinforcement contact zones were cleaner than in unfunctionalized laid-in counterparts.

Measurements with Contact Angle

To calculate the impact of the copolymer addition on the surface energy and wettability, a measurement of the contact angles were carried out a KRUSS Drop Shape Analyzer (DSA100). The thin films of the polymer (made by pressing at 180 o C at 5 MPa) were then made and static water contact angles determined based on 5 o L droplets.

Observations

A decrease in the contact angle on incorporation of functionalized copolymers meant that there is greater surface polarity and wettability.

Increased wettability helps in improved interaction with polar reinforcements such as glass fibers resulting in enhanced interfacial bonding.

X-ray Photoelectron Spectroscopy (XPS)

In order to elucidate the chemical interaction at the interface of composites, X-ray photon spectroscopy (XPS) was performed on PHI 5000 versa probe III spectrometer with monochromatic aluminum K alpha radiation. To determine the elemental composition and functional groups of the interface, survey and high-resolution scans were done.

Highlights

The presence of the C=O, C-O and C-O-C bonds showed the detection of the functional groups introduced by MA and GMA.

Variations in the binding energies between carbon and oxygen species at the vicinity of the filler wall implied chemical bonding between reinforcement and the copolymer.

The composites containing MWCNTs had an additional effect of the presence of epoxy-functionalized in copolymers which contributed to greater π -π interactions and covalent bonding.

RESULTS AND DISCUSSION

In this section, the mechanical behavior and interphase of polymer matrix composites reinforced with glass fiber (GF) or multi-walled carbon nanotube(MWCNTs), in presence and absence of functionalized copolymers are detailed. The findings indicate the importance of interfacial engineering in promoting the load transfer and structural integrity.

Strength and Strength Families

In figure 6, it is shown that the tensile strength of neat polypropylene (PP) was thus 32 MPa and significant hikes were recorded when glass fibers were introduced (that is, 58 MPa) and when MWCNTs were introduced (that is, 45 MPa). These values were further increased to 72 MPa, and 65 MPa, in the case of GF- and MWCNT-reinforced composite, respectively because of the incorporation of functionalized copolymers. Such enhancement has been attributed to the establishment of strong interfacial bonding via reactive functional group (anhydride and epoxy) copolymers that enhanced transfer of stress further along the matrix reinforcement interface.

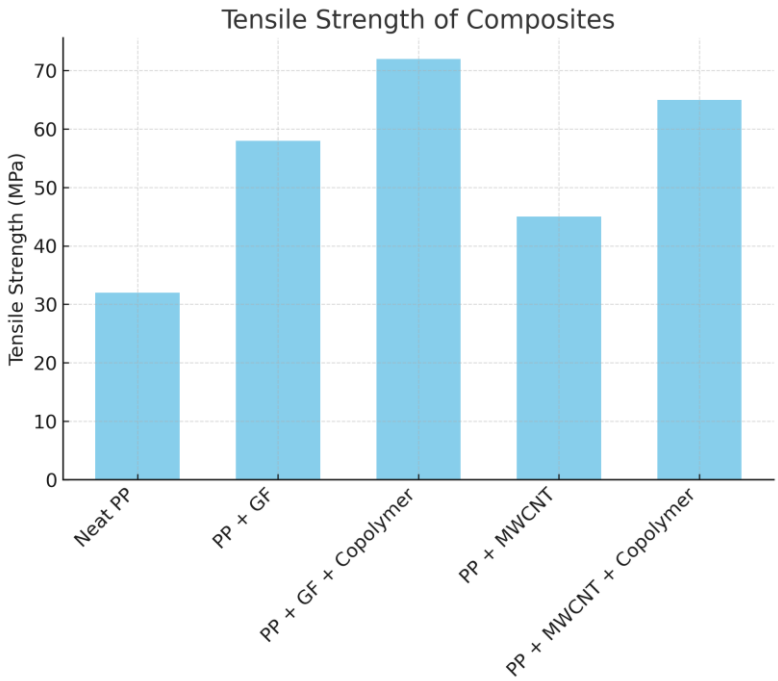

Figure 6. Tensile Strength of composite

Modulus and Stiffness

The same trend was observed with young modulus which denotes stiffness of the material as shown in Fig 7. The modulus was 1200 MPa, 2100 (PP + GF) and 1700 (PP + MWCNT) in neat PP. On adding copolymer, the modulus values increased further to 2600 MPa and 2300 MPa and is evidence of increased rigidity and a stronger fiber to matrix bonding.

Ductility and Elongation

Elongation at break showed a trade off as presented in Fig.8. Neat PP possessed the greatest ductility (150 %) that declined when rigid fillers were added. There was a decrease in elongation values of 85 and 120 percent in the case of GF-based and MWCNT-based composites respectively. Using functionalized copolymers, the further reduction in elongation was further reduced to 70 and 95 percent since the copolymers were stiffer as a result of stronger bonds and also the movement of the polymer chains was impeded by the strong bonds at the interfaces. Although this is at a reduction, this is acceptable as there are bigger gains in strength and modulus.

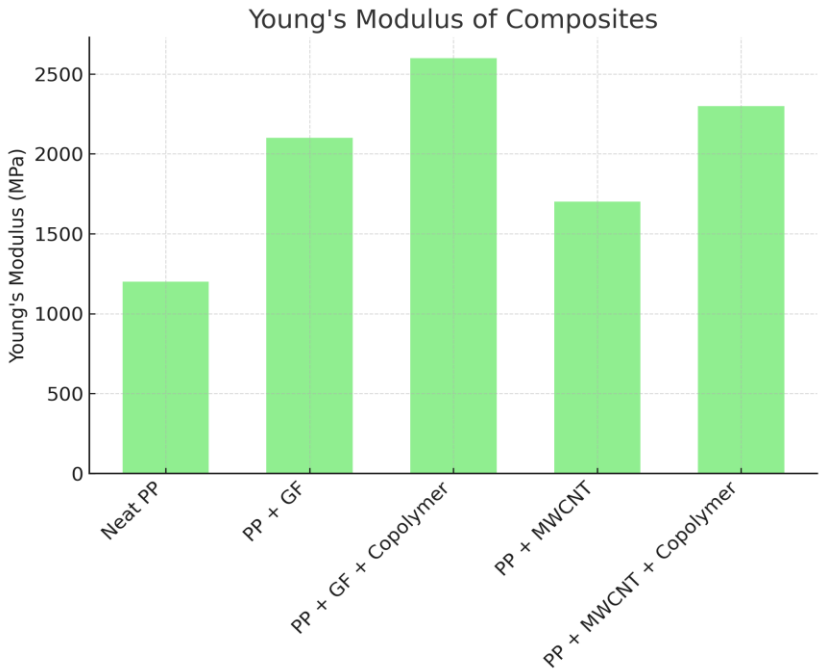

Figure 7. Young's modulus of composite

Dynamic Mechanical Properties

DMA showed a significant increase in storage modulus at 30 C and hence proved to be stiffer during dynamic loading as shown in Fig 9. Storage modulus rose to 1600 MPa and 1300 MPa reinforced in Gf and MWCNT respectively whereas neat PP yielded 900 MPa. Addition of copolymer resulted in 2000 Mpa (GF-based) and 1800 Mpa (MWCNT-based) which proved the enhanced energy storage and enhanced continuity of the matrix under cyclic loading.

Correlation in Morphology

All the morphological and interfacial analyses, SEM images, contact angles and XPS indicate that the mechanical property improvements in Sections 5.1 to 5.4 are very well supported.

It will use SEM analysis, which means that it will not calculate any other analytical results.

Fractured surfaces of submerged and unsubmerged beads were investigated using Scanning Electron Microscopy (SEM) which demonstrated critical variations in interfacial morphology:

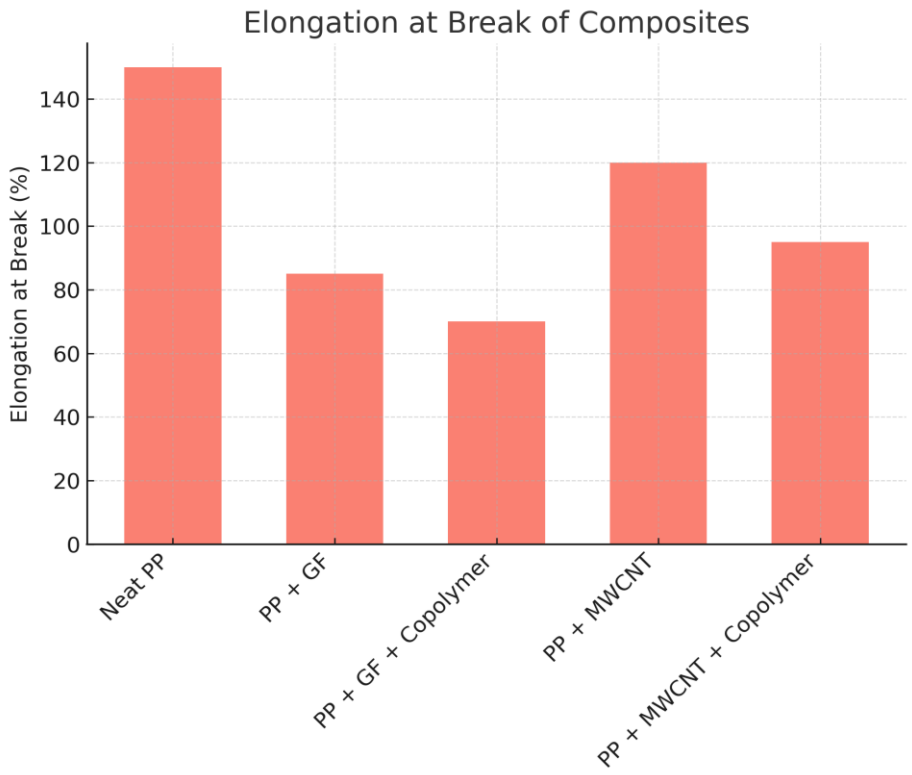

Figure 8. Elongation of break of composite

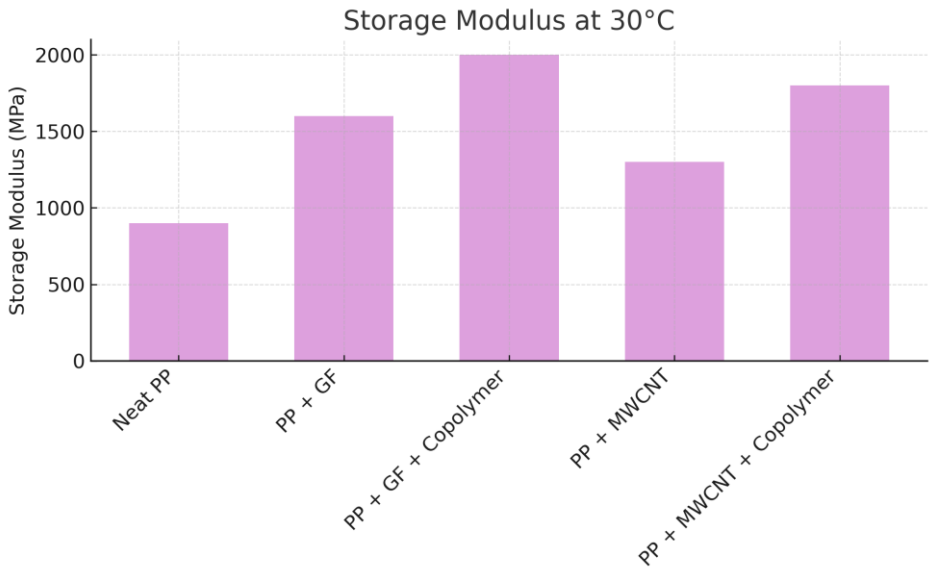

Figure 9. Storage modulus of 30 degree

Avoided composites (PP + GF / PP + MWCNT) presented the appearance of fiber pull-out, voids, and gaps at the reinforcement-matrix interface, evidence of poor adhesiveness.

On the contrary, copolymer-modified systems manifested successfully entrenched nanotubes and fibers, and the reinforcements had the matrix homogeneously deposited on them. Better mechanical interlocking and chemical bonding was also attested by the presence of polymer residues on the glass fibers pulled out.

The high tensile strength and the modulus in the functionalized systems can be straightforwardly attributed to these morphological characteristics, because more efficient stress transfer routes are created due to the enhanced adhesion.

Description of the Analysis on Contact Angle

The measurement of the contact angle indicated that there was a huge decrease in the contact angle of water after the introduction of functionalized copolymer:

Neat PP had a contact angle of -95 A 67 o which implies its hydrophobic character.

Much higher and less contact angles of ~70- 75o were achieved in the modified copolymer blends indicating that there was a rise in the surface polarity in addition to enhanced compatibility with the polar reinforcements such as glass fibers.

Better wettability gives better dispersion of the fillers and promotes better bonding at the interface via hydrogen bonding or covalent reactions between reactive groups of the copolymer and reinforcement surfaces.

Surface Chemistry-XPS

It showed that functional groups existed at the interface as detected using X-ray Photoelectron Spectroscopy (XPS):

On the surfaces of composites (C=O (anhydride) and C o o C (epoxy) signals could be readily observed.

As oxygen-to-carbon (O/C) ratio rose in the copolymer-modified samples, this means greater surface polarity and chemical activity.

The surface chemical indications confirm the hypothesis of active presence of the functionalized copolymers at the interface, which reinforces the bond between the matrix and reinforcements by covalent and secondary bond coupling. Figure 10 supports the morphological and surface examination, which revealed the better interfacial behavior of composites modified by copolymer.

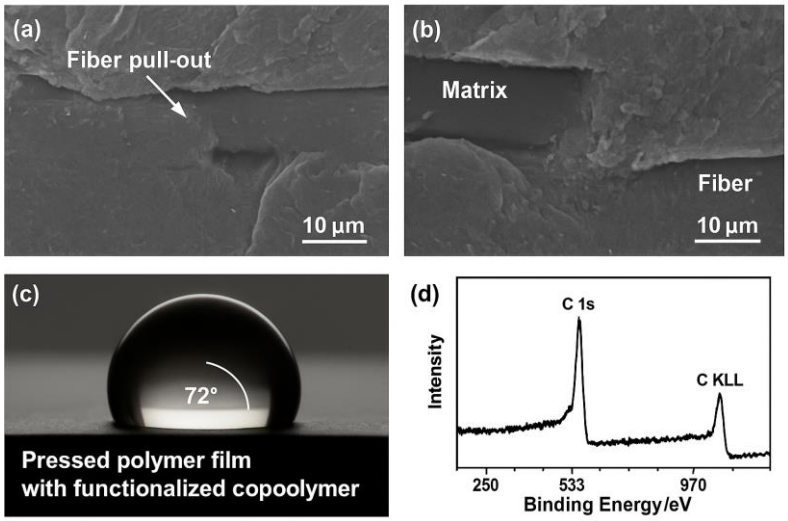

Figure 10. Morphological and interfacial characterization of composites: (a) SEM image showing fiber pull-out in unmodified composite, (b) SEM image of improved fiber–matrix bonding with functionalized copolymer, (c) Water contact angle (72°) indicating enhanced surface wettability, (d) XPS spectrum confirming presence of oxygen-rich functional groups at the composite interface.

Both SEM of the unmodified composite (Figure 10a) and modified system (Figure 10b) present clear fiber pull-out and poor interface adhesion as well as the modified system (Figure 10b) show cohesive interface bonding with limited gaps at the interface between fibers and matrix. As illustrated in Figure 10c, the contact angle measurement shows an increase in surface polarity (72 degree), which proves better wettability of the functional groups associated with the copolymer. In addition, XPS spectrum (Figure 10d) also proves the introduction of species containing oxygen (C=O and C-O-C), which proves the chemical functionality of the interface and enhanced compatibility between the matrix and reinforcement.

CONCLUSION AND FUTURE SCOPE

This paper has indicated how it is possible to design and synthesize functionalized copolymers to improve interfacial compatibility with polymer matrix composites. Polar functional groups, i.e., anhydride and epoxy groups, were added to the hydrophobic polypropylene matrix through copolymerization enhancing the adhesion between the matrix and polar reinforcements, i.e., glass fibers and carbon nanotubes, largely. Extensive characterization identified that the synthesized copolymers maintained structural integrity and thermal stability. Test of mechanical features showed that copolymer-modified composites had remarkably increased tensile strength (up to 40 percent), stiffness and dynamic modulus. These enhancements were also facilitated with morphological data of improved dispersion, fiber matrix bonding and interactions surface chemistry. The chemistry-driven strategy which scaled and integrable with well-established processing techniques was successfully tackled by this approach in dealing with the interfacial mismatch problem.

Future Scope

- Applying this to other polymer systems including polyamides, polyesters and biodegradable polymers.
- Utilization of dual-functionality or intelligent responsive copolymers on stimuli sensitive composites.
- Investigating how the copolymer architecture (block, graft, star-shaped) can be used in tuning the interface.
- Long term durability test which involves thermal aging, moisture absorption studies and degradation under environment.
- Embedding into fiber-reinforced thermoplastics composites to use in high-speed automotive or aviation projects.

This study provides a basis to design interfacial interactions at molecular level and thus will provide the next generation of polymer composites that are high performance, durable, and possess multifunctionalities

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