

# Thermoplastic-Thermosetting Merged Polyimides Derived from Furan-Maleimide

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**Abstract:** Novel thermoplastic-thermosetting merged polyimide system has been developed via Diels-Alder intermolecular reaction of bisfuran **A** namely, 2,5-bis(furan-2-ylmethyl carbamoyl)terephthalic acid with a series of bismaleimides **B**<sub>1-3</sub> containing aliphatic chain in the backbone. The intermediate Furan - maleimide Diels-Alder adducts **C**<sub>1-3</sub> were aromatized and imidized (i.e. cyclized) through carboxylic and amide groups to afford thermoplastic-thermosetting merged polyimides **D**<sub>1-3</sub>. Synthesized polyimides were characterized by elemental analysis, spectral features, number average molecular weight (Mn) and thermal analysis. Bulk polymerization was also carried out. Proof of structures was based mainly on a comparison of infrared spectra of polyimides with those of the corresponding model compound **4** prepared from 2-(furan-2-ylmethylcarbamoyl) benzoic acid in the similar way. FTIR spectral features of polyimides **D**<sub>1-3</sub> were quite identical with the model compound **4**. The 'in situ' void - free glass fiber reinforced composites **GFRC**<sub>1-3</sub> were prepared and characterized by mechanical, electrical and chemical properties. The 'in situ' produced polyimides show good adhesion to glass fibers. All the composites depicted good mechanical and electrical properties and good resistance to organic solvents and mineral acids.

**Keywords:** Diels-Alder reaction, bisfuran, bismaleimide, polyimide.

## INTRODUCTION

Polyimides have been intensively studied since the 1960s for their excellent thermal stability and high mechanical properties, along with their super chemical resistance and electrical properties [1-2]. The Diels-Alder reaction based on macromolecular chemistry has attracted much attention, particularly for providing new polyimide (PI) materials. A more elaborate approach was applied to furan-maleimide Diels-Alder polymerization. Furan heterocycle displays a pronounced dienic character, which makes it highly suitable as diene and the use of bismaleimides as dienophiles is very useful because the double bond of maleimide is very reactive towards electron rich dienes, to give a normal demand Diels-Alder cycloaddition.

The first investigation on polyimide involving a bisfuran and a bismaleimide monomer combination was reported by Tesoro and Sastri [3], who had also opened the way to a stream of publications by giving an idea of a chemical modification of Diels-Alder adduct by aromatization. Various strategies can be envisaged to synthesize or modify polymer structures by exploiting the reactivity of this heterocycle with various dienophiles by the researchers Gaina [4], Reynold [5], Sanyal [6], Peterson [7], Liu [8]. A systematic study of Diels-Alder polymerization involving furan derivatives and various bismaleimides have been carried out by Gandini [9-10]. Owing to the published

work on these topics and the long-standing interest of our laboratory in the study of polyimides via Diels-Alder reaction, a more systematic exploration has been undertaken with particular emphasis. One of the authors HSP and his co-workers have reported [11-17] the studies on polyimides through Diels-Alder polymerization of bisfuran and bismaleimide with wide structural variation.

Polyimides are of two types. Thermoplastic polyimides are usually synthesized by poly (amic acid) prepared by the reaction of an aromatic dianhydride with an aromatic diamine followed by cyclodehydration (either thermal or chemical) to the polyimide. Frequently, the final imide polymer is insoluble, infusible, and intractable hence processing of amic acid solution is used for the manufacturing of articles. Thermosetting polyimides are synthesized from unsaturated monomer e.g. bismaleimides known as thermosetting polyimides. These polyimides are processed *in-situ*. Our main concern was to synthesize novel polyimides, which possess combination of thermoplastic and thermosetting moieties by introducing bisfuran and bismaleimide segments. Introduction of bisfuran (bisamic acid/pyromellitic part) segment in adducts define as a thermoplastic portion and bismaleimide segment define as a thermosetting segment. Scheme 1 summarize our synthetic approach to the various phases of this work, viz., (i) Preparation of bisfuran **A** from pyromellitic dianhydride and furan-2-ylmethanamine (ii) Diels-Alder reaction between the **A** and **B**<sub>1-3</sub> (iii) Aromatization and imidization (i.e. cyclization) reaction of Diels-Alder adducts **C**<sub>1-3</sub> to afford Thermoplastic-thermosetting merged polyimides **D**<sub>1-3</sub>.

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The *in situ* void-free glass fiber reinforced composites **GFRC**<sub>1-3</sub> were prepared from the produced system and characterized by chemical, mechanical and electrical analysis. The details of these procedures and the results obtained are discussed below.

## MATERIAL AND METHODS

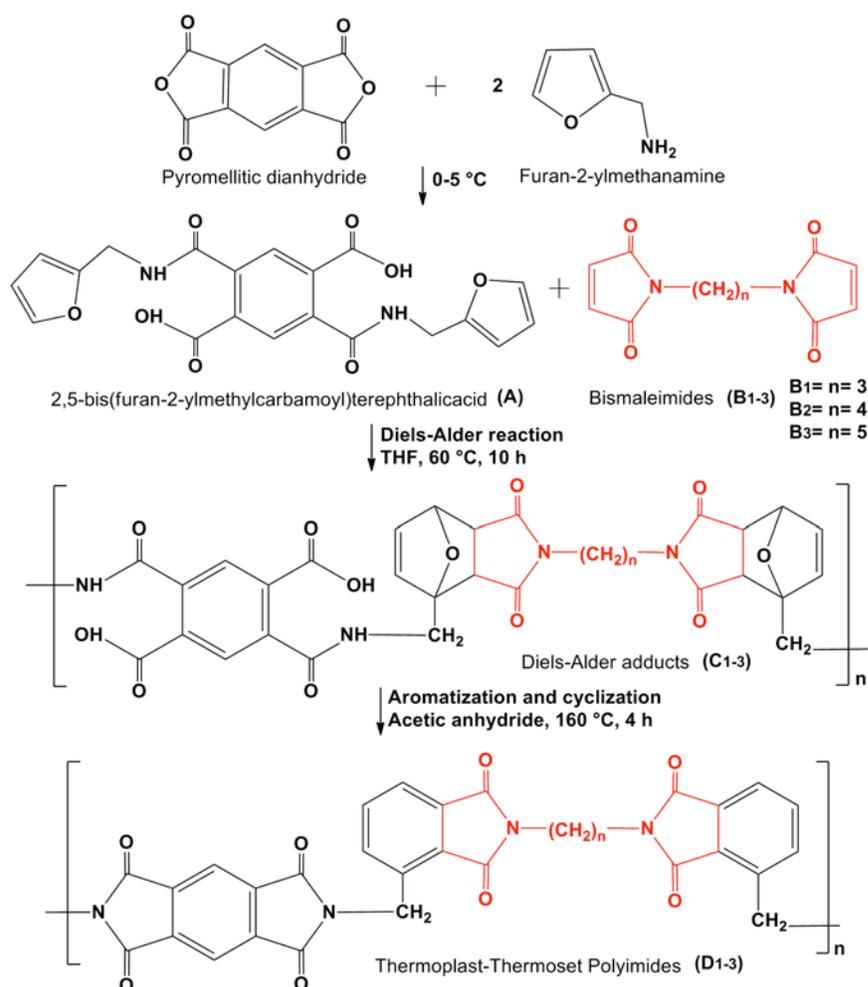
### Materials and Measurements

All chemicals and solvents used were of laboratory grade. Pyromellitic dianhydride was obtained from Fluka Analytical-Japan. Phthalic anhydride, Maleic anhydride and various diamines were obtained from local market. Furan-2-ylmethanamine was obtained from local market and redistilled before use. Solvents were dried and distilled before use according to standard procedures. Elemental analysis was carried out on a Thermofingan flash 1101EA (Italy). Infrared spectra were scanned on a Nicolet-760 FTIR spectrophotometer. <sup>1</sup>H NMR and <sup>13</sup>C NMR were

recorded on Bruker spectrophotometer (400 MHz). Number average molecular weight (M<sub>n</sub>) of polyimides was determined by non-aqueous conductometric titration by method reported [18]. Pyridine was used as suspending solvent and tetra n-butyl ammonium hydroxide in 1,4-dioxane was used as a titrant. A digital conductivity meter of Tosaniwal (India) was used for the titration. The thermal behavior of **A**, **C**<sub>1-3</sub> and **D**<sub>1-3</sub> were investigated by Thermogravimetric analysis (TGA) on Perkin Elmer TGA analyzer at heating rate of 10 Kmin<sup>-1</sup> in the temperature range of 50-700°C. As the polyimides are insoluble in common organic solvents, their viscometric properties have not been studied.

### Synthesis of Bisfuran A

**A** was synthesized by parity method [19], adding drop wise solution of furan-2-ylmethanamine (20mmol) to a stirred solution of pyromellitic dianhydride (10mmol) in 40mL of acetone and keeping the temperature of the medium close to 0-5°C for an hour



**Scheme 1:** Synthesis of Thermoplastic-thermosetting merged polyimides.

(Scheme 1), thus obtained ensuing solution was poured into ice water in which the reaction product precipitated. The final white precipitates were filtered, washed and purified by column chromatography. Yield was 65%; M. Wt. 412.35g; Decomposition temp.: 250-260 °C (uncorrected); Elemental analysis calculated for  $C_{20}H_{16}N_2O_8$ : C 58.25, H 3.91, N 6.79% Found: C 57.87, H 3.75, N 6.58%;  $^1H$  NMR ( $\delta$  ppm): 10.9 (s, 2H, -COOH), 8.9 (t, J = 5.2 Hz, 2H, -NH-CO-), 8.1 (s, 2H, Ar.H), 7.50-7.93 (m, 6H, Ar.H), 4.32 (d, J = 5.2 Hz, 4H, -CH<sub>2</sub>-);  $^{13}C$  NMR ( $\delta$  ppm): 42.24, 117.78, 119.24, 123.45, 126.10, 129.32, 132.67, 135.70, 167.12, 172.73; IR (KBr, cm<sup>-1</sup>): 3528(-OH st. acid), 3254(-NH st. amide), 3123(C-H st. furan rings), 3038(-CH st. aromatic ring), 2954(-CH st. aliphatic), 1711(C=O st. acid), 1685(O=C-NH free amide), 1627(C-C st. aromatic), 1581(COO<sup>-</sup> asym. st.), 1500(backbone st. aromatic ring), 1465(COO<sup>-</sup> sym. st.), 1249, 1175(furan in-plane CH deformation), 1039(C-O st.), 730(furan ring).

### Synthesis of Bismaleimides B<sub>1-3</sub>

Bismaleimides viz. **B**<sub>1</sub>: 1,3-bismaleimidopropane, **B**<sub>2</sub>: 1,4-bis maleimidobutane and **B**<sub>3</sub>: 1,5-bismaleimidopentane were prepared as per the method reported [20].

### Synthesis of Diels-Alder Adducts C<sub>1-3</sub>

Equimolar amount of bisfuran **A** and bismaleimide **B**<sub>1-3</sub> were dissolved in 50mL of THF. Suspension of this mixture was refluxed for 10h at 60 °C. The resultant reaction mixture was cooled and poured into a large volume of dry ether. The precipitates formed were filtered, washed and then air dried. The obtained unaromatized and uncyclized Diels-Alder adducts was designated as **C**<sub>1-3</sub> which was purified and characterized by elemental, FTIR spectral and thermal analysis.

**C**<sub>1</sub>: Yield 50%; Color light brown; Empirical Wt. 646.56g; IR (KBr, cm<sup>-1</sup>): 3530(-OH st. acid), 3252(-NH st. amide), 3121(C-H st. furan ring), 3034(-CH st. aromatic ring), 2955, 2843(-CH st. aliphatic), 1783(C=O st. asym. imide), 1730(C=O st. sym. imide), 1717(C=O st. acid), 1671(C=O st. amide), 1622(C-C st. aromatic), 1584(C=C st. furan ring), 1464(COO<sup>-</sup> st. sym.), 1243, 1162, 1061(furan in-plane CH deformation), 1040(C-O st.), 914, 873, 730(furan C-H out-of-plane deformation/C=O bending); Elemental analysis calculated for  $C_{31}H_{26}N_4O_{12}$ : C 57.59, H 4.05, N 8.67%; found: C 57.51, H 3.96, N 8.60%; Degree of

Polymerization (Dp): 7; Number average molecular weight (Mn): 4539±60.

**C**<sub>2</sub>: Yield 55%; Color light brown; Empirical Wt. 660.58g; IR (KBr, cm<sup>-1</sup>): 3523(-OH st. acid), 3242(-NH st. amide), 3115(C-H st. furan ring), 3030(-CH st. aromatic ring), 2954, 2846(-CH st. aliphatic), 1780(C=O st. asym. imide), 1736(C=O st. sym. imide), 1713(C=O st. acid), 1670(C=O st. amide), 1623(C-C st. aromatic), 1589(C=C st. furan ring), 1460(COO<sup>-</sup> st. sym.), 1240, 1163, 1062(furan in-plane CH deformation), 1045(C-O st.), 913, 870, 734(furan C-H out-of-plane deformation/C=O bending); Elemental analysis calculated for  $C_{32}H_{28}N_4O_{12}$ : C 58.18, H 4.27, N 8.48%; found: C 58.10, H 4.20, N 8.41%; Degree of Polymerization (Dp): 7; Number average molecular weight (Mn): 4630±60.

**C**<sub>3</sub>: Yield 55%; Color light brown; Empirical Wt. 674.61g; IR (KBr, cm<sup>-1</sup>): 3523(-OH st. acid), 3245(-NH st. amide), 3118(C-H st. furan ring), 3037(-CH st. aromatic ring), 2959, 2841(-CH st. aliphatic), 1783(C=O st. asym. imide), 1736(C=O st. sym. imide), 1713(C=O st. acid), 1674(C=O st. amide), 1629(C-C st. aromatic), 1586(C=C st. furan ring), 1465(COO<sup>-</sup> st. sym.), 1242, 1165, 1060(furan in-plane CH deformation), 1041(C-O st.), 911, 870, 731(furan C-H out-of-plane deformation/C=O bending); Elemental analysis calculated for  $C_{33}H_{30}N_4O_{12}$ : C 58.75, H 4.48, N 8.31%; found: C 58.67, H 4.42, N 8.23%; Degree of Polymerization (Dp): 7; Number average molecular weight (Mn): 4725±60.

### Synthesis of Polyimides D<sub>1-3</sub>

1g dried sample of **C**<sub>1-3</sub> were refluxed with 1mL of acetic anhydride [3] for 4h at 150-160 °C. The resulting solution was cooled and poured into a large volume of water. The precipitates formed were filtered, washed and then air dried. Thus obtained brown colored precipitates of aromatized and imidized polyimides designated as **D**<sub>1-3</sub> were characterized by elemental analysis, IR analysis and thermal analysis.

**D**<sub>1</sub>: Yield 45%; Color brown; Empirical Wt. 574.50g; IR (KBr, cm<sup>-1</sup>): 3042(-CH st. aromatic ring), 2960, 2854(-CH st. aliphatic), 1786(C=O st. asym. imide), 1725 (C=O st. sym. imide), 1509(backbone st. aromatic ring), 1373(imide, imide ring vibration, axial); Elemental analysis calculated for  $C_{31}H_{18}N_4O_8$ : C 64.81, H 3.16, N 9.75%; found: C 64.72, H 3.09, N 9.67%.

**D**<sub>2</sub>: Yield 45%; Color brown; Empirical Wt. 588.52g; IR (KBr, cm<sup>-1</sup>): 3045(-CH st. aromatic ring), 2955,

2853(-CH st. aliphatic), 1782(C=O st. asym. imide), 1720(C=O st. sym. imide), 1503(backbone st. aromatic ring), 1371(imide, imide ring vibration, axial); Elemental analysis calculated for  $C_{32}H_{20}N_4O_8$ : C 65.31, H 3.43, N 9.52%; found: C 65.24, H 3.37, N 9.44%.

**D<sub>3</sub>**: Yield 45%; Color brown; Empirical Wt. 602.55g; IR (KBr,  $cm^{-1}$ ): 3045(-CH st. aromatic ring), 2953, 2853(-CH st. aliphatic), 1786(C=O st. asym. imide), 1721(C=O st. sym. imide), 1503(backbone st. aromatic ring), 1376(imide, imide ring vibration, axial); Elemental analysis calculated for  $C_{33}H_{22}N_4O_8$ : C 65.78, H 3.68, N 9.30%; found: C 65.71, H 3.60, N 9.24%.

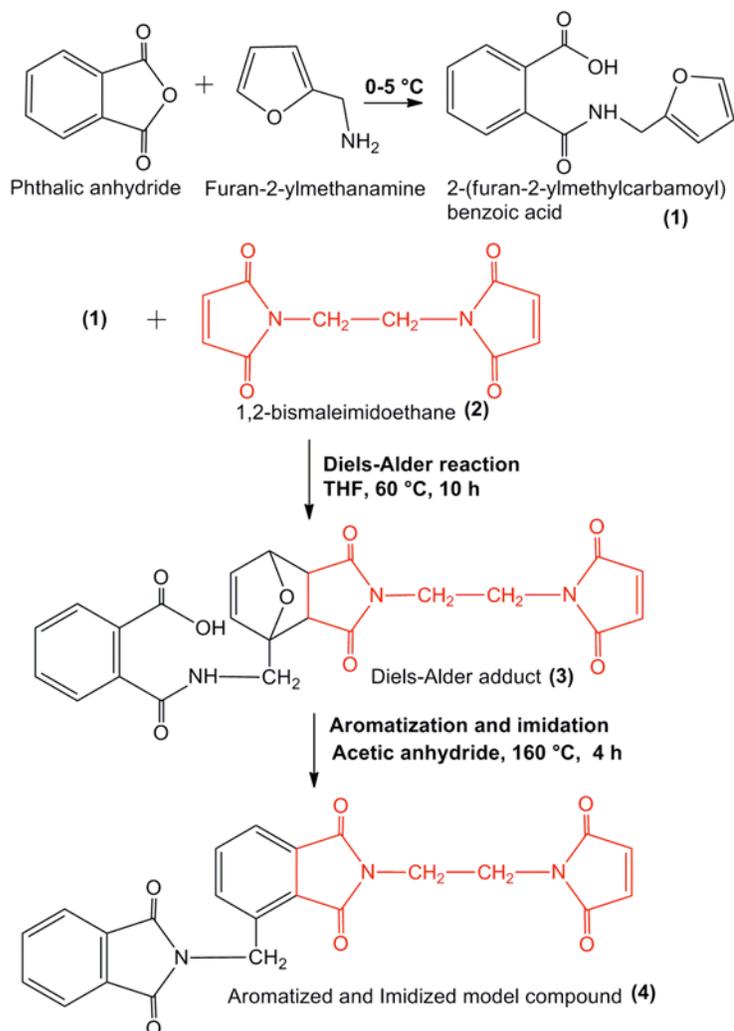
### Bulk Polymerization

Suspension mixture of **A** (10mmol), **B<sub>1-3</sub>** (10mmol) and 2 ml acetic anhydride in THF were heated at 140-150°C for 10h with vigorous agitation. The resulting products were cooled and poured into a large volume of water. The precipitates formed were filtered, washed

and then air dried. The results obtained were analogous with synthesized polyimides **D<sub>1-3</sub>**.

### Synthesis of Model Compound

A suspension of 2-(furan-2-ylmethylcarbamoyl) benzoic acid **1** (0.01mol) and ethylene bismaleimide **2** (0.01mol) in THF was refluxed for 10h at 60°C. The resultant unaromatized and unimidized product was designated as Diels-Alder adducts **3**. Unaromatized and uncyclized **3** products were refluxed at 150-160°C for 4h with 2ml of acetic anhydride to afford aromatization and imidization. The resulting solution was cooled and poured into a large volume of water, washed, filtered and then air dried. Thus obtained brown colored precipitates **4** were characterized by elemental analysis and IR analysis. Elemental Analysis Calculated for  $C_{23}H_{15}N_3O_6$ : C 64.34, H 3.52, N 9.79%, Found: C 64.07, H 3.39, N 9.61%, IR (KBr,  $cm^{-1}$ ): 3026(-CH st. aromatic ring), 2974, 2864(-CH st.



**Scheme 2:** Synthesis of model compound.

aliphatic), 1770(C=O st. asym. imide), 1735(C=O st. sym. imide), 1516(backbone st. aromatic ring), and 1376(imide, imide ring vibration, axial).

### Preparation of Glass Fiber Reinforced Composites GFRC<sub>1-3</sub>

Glass fiber reinforced composites GFRC<sub>1-3</sub> were prepared as follow: Suspensions of **A**, **B**<sub>1-3</sub> and acetic anhydride in THF were prepared on a weight basis and stirred well for 10min. The suspensions were applied with a brush to polyimide compatible glass cloth and the solvent was allowed to evaporate for 10-20min at room temperature. The dried ten prepregs so prepared were stacked one over another and placed between stainless steel plates and compressed under about 60-70psi pressure at 150°C for 10h in air circulating oven. The composites were cooled to room temperature before pressure was released. The composites were then machined to desire final dimension for various mechanical and chemical tests.

### Chemical Resistance Test

The common chemical (acids, alkali and organic solvents) resistance test for all composite samples was performed according to ASTM D543 method. This test covers the evaluation of composite materials for resistance to chemical reagents. The test includes provisions for reporting changes in weight, dimensions

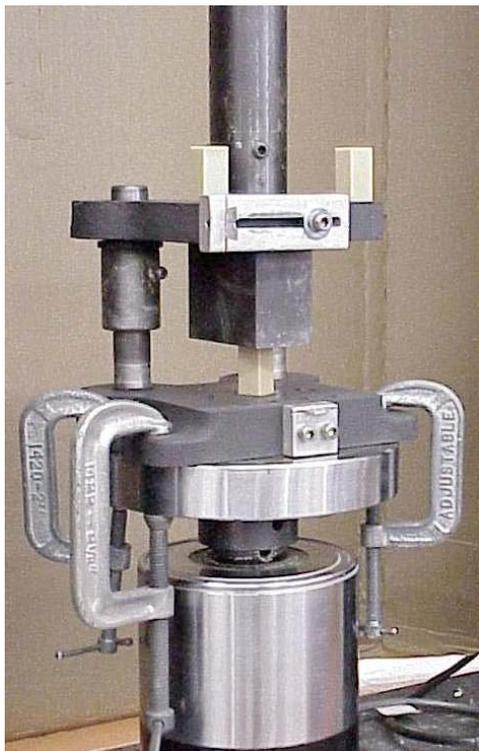


Figure 1: ASTM D695 Test Apparatus.

and appearance. The composite samples were immersed in 25% v/v H<sub>2</sub>SO<sub>4</sub>, 25% v/v HCl 25% w/v NaOH, Ethanol, acetone, DMF, and THF for 7 days at room temperature. After 7 days the samples were taken out from the reagents and analyzed for the percentage change in their thickness and weight.

### Mechanical and Electrical Tests

The compressive strength was measured according to ASTM D695 method (Figure 1). The notched izod impact strength was measured according to ASTM D256 method (Figure 2). Rockwell hardness was measured according to ASTM D785 method (Figure 3). The measurement of Flexural strength was carried out according to ASTM D790 method (Figure 4). Electrical strength measurements were carried out on a high voltage tester machine.



Figure 2: ASTM D256 Test Apparatus.

## RESULTS AND DISCUSSION

### Monomer Synthesis

To the best of our knowledge, bisfuran **A** has not been reported previously. The characterization of the reaction product provided the first unambiguous proof of the successful synthesis of bisfuran 2,5-bis(furan-2-ylmethylcarbamoyl)terephthalic acid. The FTIR spectrum of **A** showed the most relevant peaks of the furan ring and 1,2,4,5-tetra substituted benzene ring, other than the typical absorptions arising from the band at 3528cm<sup>-1</sup> and 1711cm<sup>-1</sup> for carboxylic acid and 3254cm<sup>-1</sup> and 1680cm<sup>-1</sup> for O=C-NH group [21]. In the



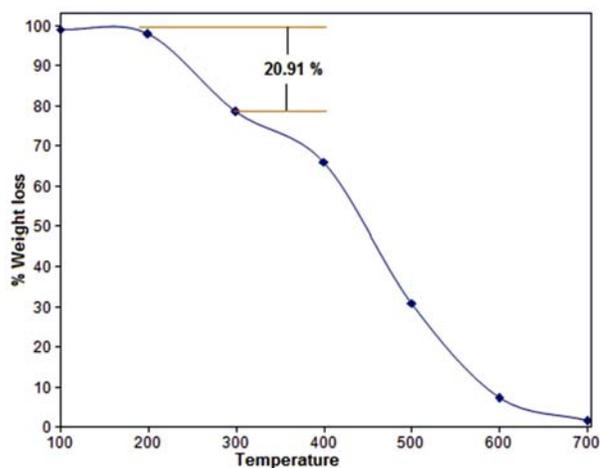
**Figure 3:** ASTM D785 Test Apparatus.



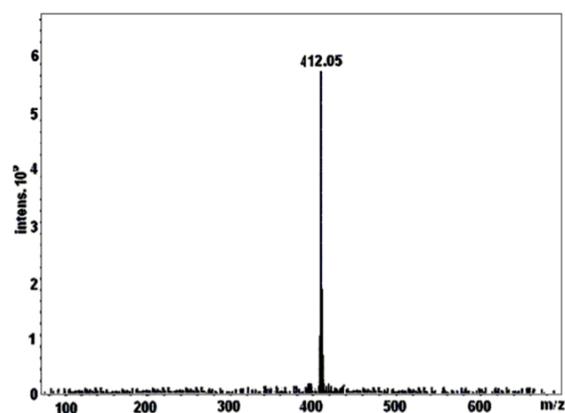
**Figure 4:** ASTM D790 Test Apparatus.

$^1\text{H}$  NMR spectroscopy, the signals in the range of 7.5–8.1ppm were ascribed to the protons of the aromatic rings. The singlet at 10.9ppm was ascribed to the protons of carboxylic –OH group and a triplet at 8.9ppm attributed to the –NH proton of amide group, which was further confirmed by  $^{13}\text{C}$  NMR values. The bisfuran **A**

started to lose weight because of thermal degradation. Thermogram of product **A** (Figure 5) indicated that, the degradation occurred into two steps. First stage of degradation starts from 180°C to 300°C might be attributed to decarboxylation of product **A**. The value of wt. loss 20.91% is consistent with the theoretical value 21.34%. The second major stage of wt. loss between 300–600°C attributed to the monomer decomposition/pyrolysis. The 3% char residue remained at 700°C. The expected structure was thus clearly verified by this spectroscopic and thermal analysis which indicated moreover the absence of any detectable impurity, particularly of the two reagents used to prepare **A**. This was additionally confirmed by LCMS data (Figure 6) and elemental analysis.



**Figure 5:** Thermogram of 2,5-bis(furan-2-ylmethylcarbamoyl) terephthalic acid **A**.



**Figure 6:** LCMS of 2,5-bis(furan-2-ylmethylcarbamoyl) terephthalic acid **A**.

### Polymer Synthesis

The Thermoplastic-thermosetting merged polyimide system has been performed by Diels-Alder

intermolecular reaction followed by aromatization and imidization simultaneously *via* cyclization. Bulk polymerization was also carried out at 150-160°C for 10h with vigorous agitation. The elemental analysis of the Diels-Alder adducts indicated that the reactions had reached good yields of cycloaddition of the bisfuran and bismaleimide rings and were consistent with their predicted structures (Scheme 1). The Degree of polymerization (Dp) for the polymers **C**<sub>1-3</sub> was estimated by non-aqueous titration was found 7-8.

The IR spectra of the Diels-Alder adducts **C**<sub>1-3</sub> showed important changes with respect to those of the initial monomers, which clearly confirmed that the Diels-Alder reaction had taken place with good yields with **B**<sub>1-3</sub>. The IR spectra of **C**<sub>1-3</sub> have prominent characteristic band of furan, amide, imide, ether and carboxylic acid groups. The bands near 3120 and 1580cm<sup>-1</sup> were attributed to furan. The bands around 3220 and 1670cm<sup>-1</sup> were attributed to amide groups, while the bands around 3530 and 1710cm<sup>-1</sup> were corresponding to -OH and -C=O group of carboxylic acid respectively. Comparison of the IR spectra of non aromatized and non cyclized **C**<sub>1-3</sub> with aromatized and imidized **D**<sub>1-3</sub> revealed discernible differences. The bands due to carboxylic acid and amide groups in the spectra of **C**<sub>1-3</sub> were almost disappeared in the spectra of **D**<sub>1-3</sub> indicated that imidization reaction had taken place very smoothly and the disappearance of the bands due to furan ring confirmed the aromatization reaction has been completed simultaneously. Proof of structure was mainly based on a comparison of infrared spectra of polyimides **D**<sub>1-3</sub> with those of the corresponding model compound **4**. IR spectral features of **D**<sub>1-3</sub> were quite identical with the model compound **4**. These features confirmed the predicted structure of aromatized and imidized **D**<sub>1-3</sub>.

The thermal stability of the **C**<sub>1-3</sub> and **D**<sub>1-3</sub> were evaluated from their TG curves. The **C**<sub>1-3</sub> undergoes two stages of mass loss. The first stage corresponded to the decarboxylation of the polymer in the temperature range of 180°C to 300°C. The values of wt. loss observed in **C**<sub>1-3</sub> (Figure 7) were quite consistent with the theoretical values. It indicated that the Diels-Alder reaction was completed as expected. The process of the second stage above 300°C was due to polymer pyrolysis. The char residue left at 700°C was in the range of 4-6% for **C**<sub>1-3</sub>. Very rapid rate of weight loss was observed between 400°C to 600°C. Whereas in the case of **D**<sub>1-3</sub> started their decomposition after 400°C (Figure 8) depending upon the nature of polyimides. Almost all polymer samples showed 50% weight loss in

the range of 500°C to 600°C. Complete weight loss (around 95–96%) was observed at about 700°C. The aromatized and imidized samples **D**<sub>1-3</sub> were more thermally stable as their pre polymers **C**<sub>1-3</sub> have nature of decarboxylation. Thus **D**<sub>1-3</sub> started their degradation at slightly higher temperature as compared to non-aromatized samples **C**<sub>1-3</sub>. Comparison of thermal stability of all polyimides revealed the following order of stability: **B**<sub>1</sub> > **B**<sub>2</sub> > **B**<sub>3</sub>. The reported thermal data of most of commercial polyimides were determined in N<sub>2</sub>. The present study was carried out in air. So it was worthwhile to compare, but it could be concluded that the produced polyimides were quite thermally stable and competent to commercial polyimides.

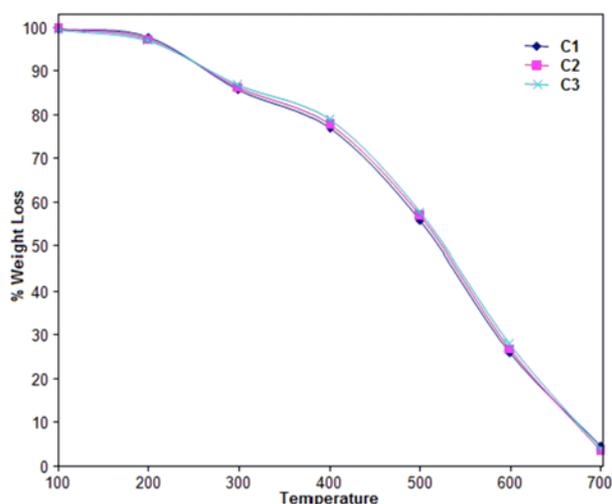


Figure 7: Thermogram of **C**<sub>1-3</sub>.

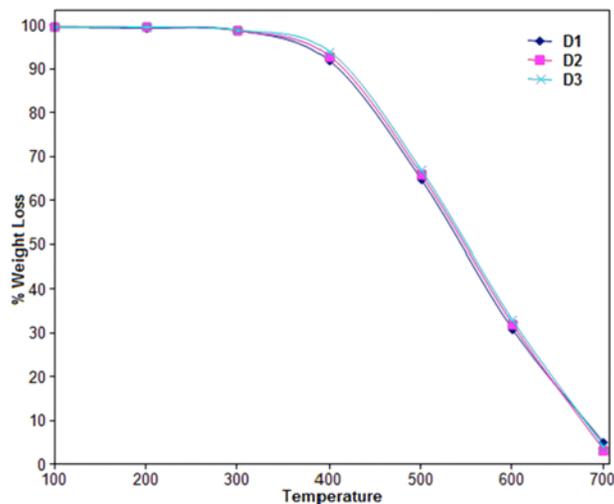


Figure 8: Thermogram of **D**<sub>1-3</sub>.

### Composite Characterization

The 'in situ' void-free glass fiber reinforced dark brown colored composites were prepared from

**Table 1: Chemical Resistance of Glass fiber Reinforced Composites GFRC<sub>1-3</sub>**

Composite <sup>a</sup>	GFRC1 % Change		GFRC2 % Change		GFRC3 % Change	
	Thickness	Wt.	Thickness	Wt.	Thickness	Wt.
25% H <sub>2</sub> SO <sub>4</sub>	1.1	1.2	1.2	1.2	1.2	1.1
25% HCl	1.0	1.0	1.0	1.1	1.1	1.1
25% NaOH	1.2	1.1	1.2	1.1	1.2	1.3
Ethanol	0.8	1.0	0.8	1.0	0.9	1.1
Acetone	0.9	1.0	0.8	1.0	1.1	1.2
DMF	1.7	1.9	1.7	1.9	1.7	1.8
THF	1.1	1.1	1.1	1.2	1.0	1.1

<sup>a</sup>Condition: Reinforcement-E type glass cloth, Plain weave, 10 mm, 10 layers, Resin Content: 40±2%, Weight % molar ratio: - A: B<sub>1-3</sub> = 1:1(mol/mol), Curing temperature = 150±10 °C, Curing time = 10h, Curing pressure = 60-70psi, Composite size: 25mm × 25mm × 3mm.

**Table 2: Mechanical and Electrical Property of Glass Fiber Reinforced Composites GFRC<sub>1-3</sub>**

Composite	Flexural strength (MPa)	Compressive strength (MPa)	Notched Impact strength (MPa)	Rockwell hardness (MPa)	Electrical strength in air (kv/mm)
GFRC1	280	230	250	126	15.9
GFRC2	270	228	245	125	16.6
GFRC3	275	225	240	124	17.9

produced system and characterized. The percentage weight loss in composite samples is summarized in Table 1. All the samples showed excellent acid, alkali and organic solvent resistance and no significant change in the physical appearance. However, the concentrated alkali cause changes of about 1.1-1.3% and DMF cause changes of 1.8-1.9% in their thickness and weight. Mechanical tests were performed using their specimens. The results of Mechanical test (Table 2) revealed that all the composites have good mechanical properties especially due to presence of bismaleimides. The overall trend of the mechanical properties of the prepared polyimide glass fiber reinforced composites decreases as follows: **GFRC<sub>1</sub> > GFRC<sub>2</sub> > GFRC<sub>3</sub>**. This may be attributed to a certain increase in the rigidity of the bismaleimide component. The electric strength of all the composites is in the range of 15.9–17.9kV/mm. As there is no wide change in electrical strength, no attempt was made to interpret these data.

## CONCLUSIONS

A novel polyimides containing combination of both thermoplast and thermoset in merged segment system have been successfully developed through Diels-Alder reaction of 2,5-bis(furan-2-ylmethyl carbamoyl)

terephthalic acid with different bismaleimides, followed by aromatization and imidization reaction. The 'in situ' produced polyimides showed good adhesion to glass fibers. Void-free glass fiber reinforced composites have good mechanical and electrical properties and good resistance to organic solvents and mineral acid.

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Received on 22-10-2012

Accepted on 03-12-2012

Published on 31-12-2012

DOI: <http://dx.doi.org/10.6000/1929-5995.2012.01.02.3>