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# A Simple Single-Step Synthesis of *Bis*(indolyl)methane using Polyaniline-Supported Graphene Oxide Nanocomposites as Catalysts

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A novel approach for synthesizing *bis*(indolyl)methane derivatives (**3a-k**) through the reaction of aromatic aldehyde and 5-bromo indole in ethanol is presented. The reaction utilizes a heterogeneous catalyst composed of polyaniline supported graphene oxide, which offers environmental benefits. Advantages of the approach over previously described methods include a shorter reaction time, milder reaction conditions, and a reusable catalyst. Furthermore, this synthetic route aligns with the principles of green chemistry by fulfilling various important requirements. The synthesized catalysts, consisting of heterogeneous polyaniline supported graphene oxide, were thoroughly characterized using SEM, IR and XRD techniques.

**Keywords: Polyaniline supported graphene oxide, Green synthesis, *Bis*(indolyl)methane, Reusability.**

## INTRODUCTION

In recent years, the *bis*(indolyl)methane moiety has been extensively studied in both natural products and synthetic compounds with significant biological and medicinal importance. This indole heterocyclic compound is commonly found in numerous natural compounds. *Bis*(indolyl)methane and its derivatives play a crucial role in organic compounds, exhibiting a wide range of pharmaceutical and biological activities including anticancer [1], antibacterial, antioxidant [2], anti-HIV [3], antimicrobial, anti-inflammatory [4], antibiotic, antiviral, anti-neurodegenerative and analgesic [5] properties. Moreover, this heterocyclic compound finds utility as an insecticide, fungicide, pigment and insecticide [6,7].

The attention towards *bis*(indolyl)methane has significantly increased in recent years due to its presence in various natural products and its importance in agriculture, biomedicine and plant growth. Significantly, alkaloids, including arsindoline A and B, which have been extracted from marine bacteria, have demonstrated considerable anti-tumor properties when tested against several types of cancer cells [8]. Vibrindole A demonstrates diverse antibacterial activities [9], while arundin is employed in cancer chemotherapy. Streptindole, a genotoxic metabolite of human intestinal bacteria, is also associated with *bis*(indolyl)-methane [10]. Therefore, *bis*(indolyl)methane is a versatile natural product that plays a crucial role in agriculture, biomedicine and plant growth. The synthesis of *bis*(indolyl)methane through multicomponent reactions has garnered significant attention due to its one-pot green chemical method, solvent-less synthesis, eco-friendly feasibility, low hazards, minimal energy consumption, reduced reagents and environ-mentally friendly reaction conditions. Furthermore, *bis*(indolyl)methane finds application in highly selective colorimetric and ratiometric fluorescent molecular chemosensors as well as in cancer chemotherapy [11].

Various methods have been reported for the synthesis of *bis*(indolyl)methanes over the years. Typically, this compound can be obtained by reacting an indole or substituted indole with an aromatic or aliphatic aldehyde using a Lewis acid, such as $FeCl_3 \cdot 6H_2O$ [12], xanthan perchloric acid [13], ascorbic acid [14] or micron particulate AlN/Al [15]. Heteropoly-11-tungsto-1-vanadophosphoric acid [16], 12-tungstosilicic acid [17], taurine [18], a zirconium(IV) complex of N,O type *p-tert*-butylcalix[4]-arene [19], Ir-catalyst supported on a substrate [20], function-alized ionic liquid [21], chiral phosphoric acid catalyst [22], nano-$Al_2O_3$ [23], Seralite SRC-120 resin [24], tetramethyl guanidinium chlorosulphonate [25], pentafluorophenylamm-

onium triflate [26], ethylammonium nitrate [27], oxalate-capped iron nanomaterials [28], nanocopper ferrite catalyst [29] and a propylsulfonic acid-anchored isocyanurate based periodic mesoporous organosilica [30] are some examples of the compounds or materials used in various studies. Currently, there is growing interest in the utilization of organic solvents as alternative catalysts supported by polyaniline and graphene oxide. This method has proven effective in the synthesis of *bis*-indole and its derivatives. The use of polyaniline-supported metal oxide catalysts offers a straightforward, appealing and environmentally friendly one-pot approach for the synthesis of *bis*(indolyl)-methanes, accommodating a broad range of functional groups. Some selected biologically active *bis*(indolyl)methane are shown in Fig. 1.

In this study, an efficient and convenient approach is presented for synthesizing *bis*(indolyl)methanes (**3a-k**) by reacting 5-bromoindole and aromtic aldehyde using a heterogeneous catalyst composed of polyaniline supported on graphene oxide.

## EXPERIMENTAL

**Synthesis of graphene oxide (GO):** Graphene oxide (GO) synthesized by modified Hummer method. Generally, flake graphite (5 g), $KMnO_4$ (3 g) and $K_2FeO_4$ (2 g) as oxidant and boric acid (0.01 g) as stabilizer were first dispersed in a 50 mL of conc. $H_2SO_4$ in a vessel and stirred for 1.5 h at less than 5 ºC. After 3 h of stirring at this temperature, 2 g $KMnO_4$ were added, and the vessel was placed in a water bath heated to 35 ºC to complete the oxidation process. It was diluted with slowly addition of 100 mL water at 95 ºC and held for constant for 15 min and the colour changed to brown suspension. Finally, this dilute brown suspension reacted with 10 mL $H_2O_2$ to terminate the reaction, which turns the solution into yellow colour and washed it with 1 mol/L HCl and followed by deionized water for several times. The powder form of graphene oxide was obtained by vacuum drying at room temperature following the filtration process.

**Synthesis of PANI-GO nanocomposite:** The synthesis of polyaniline supported graphene oxide (PANI-GO) was conducted through chemical polymerization. Initially, 1.5 g of aniline hydrochloride was added to 50 mL of double distilled water and agitated for 20 min to obtain a homogeneous mixture. The mixture was then placed in an ice bath at 100 ºC for 2 h, resulting in solution (A). In solution (A), 2 wt.% of graphene oxide nanoparticles were dispersed. Next, 2.5 g of ammonium persulfate was dissolved in 15 mL of distilled water and kept in an ice bath for 2 h, yielding solution (B). Solution (B) was added dropwise to solution (A) over a period of 3 h. Throughout this process, the reaction mixture composed of (A) and (B) was continuously stirred. Upon the addition of ammonium persulfate, the reaction mixture rapidly turned greenish black, indicating that graphene oxide (GO) expedited the synthesis of PANI/GO. The resulting greenish black powder obtained from the reaction was utilized for further characterization.

**General procedure for synthesis of *bis*(indolyl)methane (3a-k):** A catalyst comprising 10 mol% polyaniline supported on GO was introduced to a solution containing aromatic aldehyde (1 mmol) and 5-bromoindole (2 mmol) in ethanol (5 mL). Subsequently, the mixture was stirred at 60 ºC while the progress of the reaction was monitored using TLC. Once the reaction reached completion, the solution was taken off the heat, poured onto crushed ice and the resulting solid products were separated *via* simple filtration. The solid products (**3a-k**) were then washed with ice-cold water (**Scheme-I**) and then recrystallized with 99% ethanol.

**3,3′-((4-Chlorophenyl)methylene)*bis*(5-bromo-1*H*-indole) (3a):** Yield: 87%; m.p.: 70-72 ºC; Reflux time: 2 h. Mass *m/z*: 513, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.00 (s, 2H), 7.46 (s, 2H), 7.29-7.28 (d, *J* = 2.9 Hz, 4H), 7.26-7.25 (m, 4H), 6.62 (s, *J* = 2.4 Hz, 2H), 5.73 (s, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ ppm: 141.77, 135.50, 132.39, 130.05, 128.78, 128.63, 125.31, 124.94, 122.31, 118.70, 112.97, 112.87, 39.46.

**3-(*Bis*(5-bromo-1*H*-indol-3-yl)methyl)phenol (3b):** Yield: 85%; m.p.: 122-124 ºC; Reflux time: 1 h. Mass *m/z*: 496, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.01 (s, 2H), 7.48 (s, 2H), 7.26-7.24 (d, *J* = 1.8 Hz, 4H), 7.22-7.18 (m, 4H), 6.90 (dd, *J* = 2.5, 1.0 Hz, 2H), 6.87 (s, 1H). 5.70 (s, 1H). $^{13}$C NMR

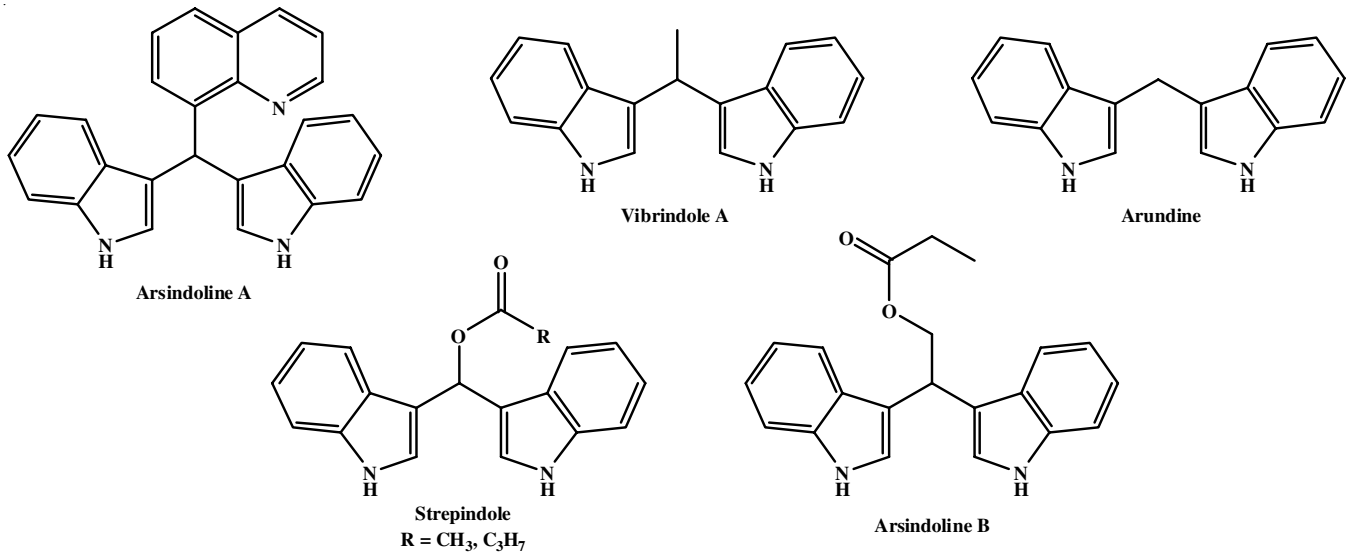
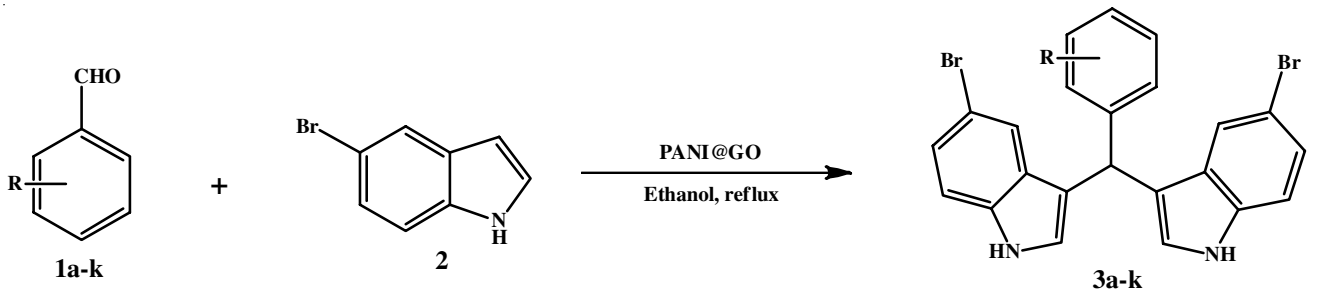



Fig. 1. Some selected biologically active *bis*(indolyl)methane




**Scheme-I:** Synthesis of 3,3'-((substituted phenyl)methylene)*bis*(5-bromo-1*H*-indole); Reaction condition (**3a**): 4-chlorobenzaldehyde (**1**) (1 mmol), 5-bromoindole (**2a**) (1 mmol), ethanol, catalyst reflux 2-3 h

(101 MHz, $CDCl_3$) δ ppm: 150.84, 146.77, 135.34, 129.42, 128.25, 125.43, 124.85, 123.87, 121.92, 117.43, 113.04, 112.89, 39.84.

**3,3′-((4-Bromophenyl)methylene)*bis*(5-bromo-1*H*-indole) (3c):** Yield: 80%; m.p.: 110-112 ºC; Reflux time: 1.5 h. Mass *m/z*: 559, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.01 (s, 2H), 7.47 (s, 2H), 7.30-7.28 (d, *J* = 2.9 Hz, 4H), 7.27-7.22 (m, 4H), 6.61 (s, *J* = 2.4 Hz, 2H), 5.74 (s, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ ppm: 142.31, 135.52, 131.74, 130.48, 128.63, 125.34, 124.95, 122.32, 120.54, 118.62, 113.00, 112.85, 39.54.

**3,3′-(Phenylmethylene)*bis*(5-bromo-1*H*-indole) (3d):** Yield: 94%; m.p.: 126-128 ºC; Reflux time: 1 h. Mass *m/z*: 480, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.02 (s, 2H), 7.50 (s, 2H), 7.32 (d, *J* = 2.9 Hz, 3H), 7.29-7.26 (m, 5H), 6.67 (d, *J* = 2.4 Hz, 2H), 5.78 (s, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ ppm: 143.04, 135.34, 128.66, 128.55, 128.46, 126.55, 124.99, 124.78, 122.29, 119.07, 112.69, 112.60, 39.89.

**4-(*Bis*(5-bromo-1*H*-indol-3-yl)methyl)phenol (3e):** Yield: 82%; m.p.: 195-197ºC; Reflux time: 1.5 h. Mass *m/z*: 496, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 7.96 (s, 2H), 7.47 (s, 2H), 7.26-7.24 (d, *J* = 1.8 Hz, 4H), 7.23-7.21 (m, 4H), 7.15 (dd, *J* = 2.5, 1.0 Hz, 2H), 6.62 (s, 1H). 5.68 (s, 1H). $^{13}$C NMR (101 MHz, $CDCl_3$) δ ppm: 154.21, 135.60, 135.13, 129.85, 128.80, 125.13, 124.87, 122.49, 119.50, 115.45, 112.82, 112.77, 39.22.

**3,3′-((4-Fluorophenyl)methylene)*bis*(5-bromo-1*H*-indole) (3f):** Yield: 90%; m.p.: 74-76 ºC; Reflux time: 1 h. Mass *m/z*: 498, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.01 (s, 2H), 7.70 (s, 2H), 7.26-7.24 (d, *J* = 1.8 Hz, 4H), 7.23-7.21 (m, 4H), 7.15 (dd, *J* = 2.5, 1.0 Hz, 2H), 6.32 (s, 1H). 5.80 (s, 1H). $^{13}$C NMR (101 MHz, $CDCl_3$) δ ppm: 162.97, 160.54, 138.90, 135.53, 130.15, 128.68, 125.27, 124.89, 122.38, 119.08, 115.31, 112.93, 112.83, 39.33.

**3,3′-((3,4-Dimethoxyphenyl)methylene)*bis*(5-bromo-1*H*-indole) (3g):** Yield: 80%; m.p.: 196-198 ºC; Reflux time: 2 h. Mass *m/z*: 541, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.05 (s, 2H), 7.51 (d, *J* = 1.6 Hz, 2H), 7.29 (d, *J* = 3.4 Hz, 2H), 7.27-7.26 (m, 3H), 6.80 (d, *J* = 1.8 Hz, 2H), 6.69-6.64 (m, 2H), 5.72 (s, 1H), 3.89 (s, 3H), 3.80 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ ppm: 148.82, 147.56, 135.71, 135.36, 128.65, 124.97, 124.74, 122.31, 120.43, 119.23, 112.67, 112.63, 111.95, 111.00, 55.87, 39.51.

**4-(*Bis*(5-bromo-1*H*-indol-3-yl)methyl)-2-methoxyphenol (3h):** Yield: 83%; m.p.: 111-113 ºC; Reflux time: 1 h. Mass *m/z*: 526, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.03 (s, 2H), 7.51 (d, *J* = 1.6 Hz, 2H), 7.28 (s, 1H), 7.27-7.26 (m, 3H), 6.86 (d, *J* = 8.2 Hz, 2H), 6.76 (dd, *J* = 8.1, 2.0 Hz, 1H), 6.67 (d, *J* = 2.4 Hz, 2H), 5.70 (s, 1H), 5.57 (s, 1H), 3.81 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ ppm: 146.48, 144.12, 135.35, 135.11, 128.64, 124.96, 124.74, 122.30, 121.14, 119.32, 114.19, 112.66, 112.62, 111.20, 55.92, 39.57.

**5-(*Bis*(5-bromo-1*H*-indol-3-yl)methyl)-2-methoxyphenol (3i):** Yield: 82%; m.p.: 110-112 ºC; Reflux time: 1 h. Mass *m/z*: 526, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.04 (s, 2H), 7.55-7.48 (m, 2H), 7.31-7.19 (m, 5H), 6.82 (d, *J* = 21.1 Hz, 3H), 6.61 (d, *J* = 2.4 Hz, 2H), 5.66 (s, 1H), 3.88 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ ppm: 145.35, 145.19, 136.51, 135.33, 128.63, 124.86, 124.72, 122.23, 120.05, 119.12, 114.77, 112.64, 112.58, 110.55, 55.97, 39.24.

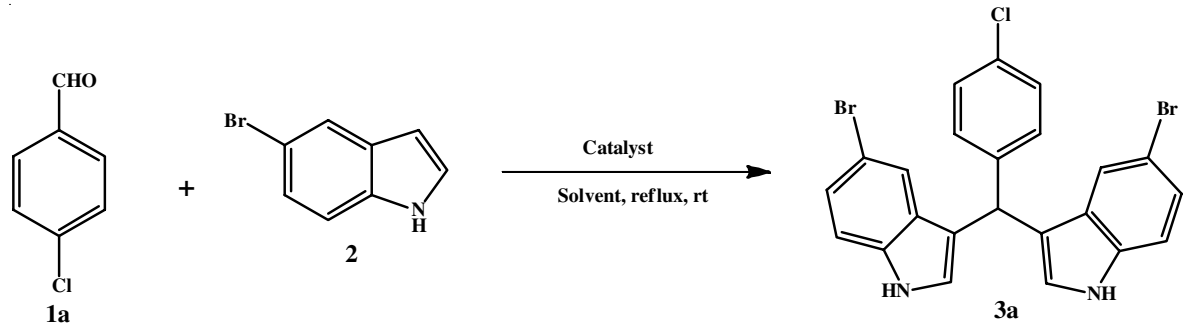
**3,3′-((4-Nitrophenyl)methylene)*bis*(5-bromo-1*H*-indole) (3j):** Yield: 92%; m.p.: 230-232 ºC; Reflux time: 1.5 h. Mass *m/z*: 525, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.09 (s, 2H), 7.52 (d, *J* = 1.8 Hz, 2H), 7.31-7.26 (m, 5H), 6.90 (t, *J* = 7.4 Hz, 2H), 6.80-6.71 (m, 2H), 5.96 (s, 1H), 5.22 (s, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ ppm: 153.94, 135.51, 129.74, 128.50, 128.27, 125.35, 124.75, 122.29, 121.04, 116.99, 116.53, 112.96, 112.76, 34.81.

**2-(*Bis*(5-bromo-1*H*-indol-3-yl)methyl)phenol (3k):** Yield: 85%; m.p.: 240-242 ºC; Reflux time: 1.5 h. Mass *m/z*: 496, $^1$H NMR (400 MHz, $CDCl_3$) δ ppm: 8.21-8.17 (m, 2H), 8.15 (s, 2H), 7.48 (d, *J* = 2.0 Hz, 1H), 7.46 (d, *J* = 1.8 Hz, 3H), 7.33-7.29 (m, 4H), 6.68 (dd, *J* = 2.5, 1.0 Hz, 2H), 5.88 (s, 1H). $^{13}$C NMR (101 MHz, $CDCl_3$) δ ppm: 150.84, 146.77, 135.34, 129.42, 128.25, 125.43, 124.85, 123.87, 121.92, 117.43, 113.04, 112.89, 39.84.

## RESULTS AND DISCUSSION

**Effect of solvent and catalyst:** In present work, *bis*-(indolyl)methane was synthesized by reacting 4-chlorobenzaldehyde (**1a**) (1 mmol) and 5-bromoindole (**2**) (1 mmol) using different catalysts and different solvents at reflux temperature (**Scheme-II**, Table-1). Among these, the catalyst observed that polyaniline supported graphene oxide exhibited high catalytic activity and gave high yield% of product **3a**. Similarly, it was found that ethanol solvent has the potential to produce the maximum possible yield (Table-2).

Table-3 shows a series of reactions under different weights of tested catalysts. In control reaction, no catalyst is utilized and no change in the reactant is observed. The polyaniline supported graphene oxide is verified as catalyst with load 20 and 60 mg grounded with 4-chlorobenzaldehyde and 5-bromoindole with




**Scheme-II:** Synthesis of 3,3'-((4-chlorophenyl)methylene)*bis*(5-bromo-1*H*-indole) (BIMs); Reaction condition (**3a**): 4-chlorobenzaldehyde (**1**) (1 mmol), 5-bromoindole (**2a**) (1 mmol), ethanol, catalyst, reflux 2-3 h

TABLE-1
COMPARISON OF THE EFFICIENCY OF PANI@GO CATALYST FOR THE SYNTHESIS OF BIMs

| Catalyst | Solvent | Time (h) | Temp. (°C) | Yield (%) | Ref. |
|---|---|---|---|---|---|
| Taurine | $CH_2Cl_2$ | 24 | 23 | 52 | [11] |
| Fe(ox)-$Fe_3O_4$ | Water | 6 | 70 | 68 | [21] |
| Supported Ir catalyst | Methanol | 4 | 150 | 61 | [13] |
| $ZrO_2$ | Ethanol | 4 | 80 | 66 | [12] |
| $CuFe_2O_4$ | Methanol | 60 | 62 | 30 | [22] |
| PANI@GO | Ethanol | 1 | 80 | 94 | Present work |

Reaction condition (**3a**): 4-Chlorobenzaldehyde (**1**) (1 mmol), 5-bromoindole (**2a**) (1 mmol), ethanol, catalyst reflux 1-2 h

TABLE-2
SCREENING OF THE SOLVENT FOR THE SYNTHESIS OF BIM (**3a**)

| Entry | Method used | Solvent | Time (h) | Temp. (°C) | Yield (%) |
|---|---|---|---|---|---|
| 1 | Stirring | Water | 5.0 | rt | 84 |
| 2 | Reflux | Water | 3.0 | 70 | 86 |
| 3 | Stirring | Hexane | 4.0 | rt | 87 |
| 4 | Reflux | Hexane | 2.0 | 80 | 90 |
| 5 | Stirring | DMF | 4.0 | rt | 88 |
| 6 | Reflux | DMF | 2.0 | 80 | 88 |
| 7 | Stirring | Ethanol | 3.5 | rt | 89 |
| 8 | Reflux | Ethanol | 1.0 | 80 | 94 |

Reaction condition (**3a**): 4-Chlorobenzaldehyde (**1**) (1 mmol), 5-bromoindole (**2a**) (1 mmol), ethanol, catalyst reflux 1-2 h

TABLE-3
EFFECT OF CATALYST LOADING ON THE SYNTHESIS

| Entry | Catalyst (mg) (PANI@GO) | Time (h) | Yield (%) |
|---|---|---|---|
| 1 | 0 | 6.0 | – |
| 2 | 20 | 5.0 | 60 |
| 3 | 35 | 4.5 | 65 |
| 4 | 50 | 4.0 | 70 |
| 5 | 65 | 3.0 | 73 |
| 6 | 80 | 2.5 | 75 |
| 7 | 90 | 2.0 | 80 |
| 8 | 100 | 1.0 | 94 |

Reaction condition (**3a**): 4-Chlorobenzaldehyde (**1a**) (1 mmol), 5-bromoindole (**2**) (1 mmol), ethanol, catalyst reflux 1-2 h

4 and 3 h, respectively, obtained 60% and 65% yield. The amount of polyaniline supported graphene oxide was increased to 100 mg in order to increase the product yield. The same yield was observed with increasing amount of catalyst (Table-3).

**Recycling of catalyst:** It was discovered that PANI-GO exhibited recyclability in the synthesis of *bis*(indolyl)methane (**3a**) from 4-chlorobenzaldehyde (**1a**) and 5-bromoindole (**2**) using ethanol as solvent and refluxed the reaction mixture for 1 h. The catalyst consisting of polyaniline supported on graphene oxide was successfully recovered through filtration and could be reused multiple times without any loss in its effectiveness (Table-4).

TABLE-4
RECYCLING OF PANI-GO FOR THE SYNTHESIS OF BIM (**3a**)

| Run | Catalyst recovery | Time (h) | Yield |
|---|---|---|---|
| 1 | 94 | 1 | 94.5 |
| 2 | 94 | 1 | 94.0 |
| 3 | 93 | 1 | 93.5 |
| 4 | 92 | 1 | 91.5 |
| 5 | 91 | 1 | 91.0 |

## Characterization of catalyst

**X-ray diffraction study:** Fig. 2 displays the XRD patterns of three materials, PANI, GO and PANI/GO-2%. The XRD analysis of synthesized GO reveals a distinct, sharp peak at approximately $2\theta = 12.6^\circ$ (001), indicating its crystalline nature. Additionally, the peak at $2\theta = 26.5^\circ$ (002) suggests the existence of unoxidized graphene, while the peak at 42.5° corresponds to a carbon peak. Nevertheless, the incorporation of polyaniline as base material in the modification of GO leads to a significant reduction in the intensity of these three distinct peaks. This reduction can be attributed to the utilization of a lesser quantity of GO during the modification procedure. This decrease in intensity signifies a reduction in crystallinity and is accompanied by prominent peaks at 15°, 20° and 25.6°, which indicate the formation of a PANI/GO-2% nanocomposite as shown in Fig. 2c. These results suggest that GO is essential for the stability of crystalline PANI chains embedded in an amorphous substrate.

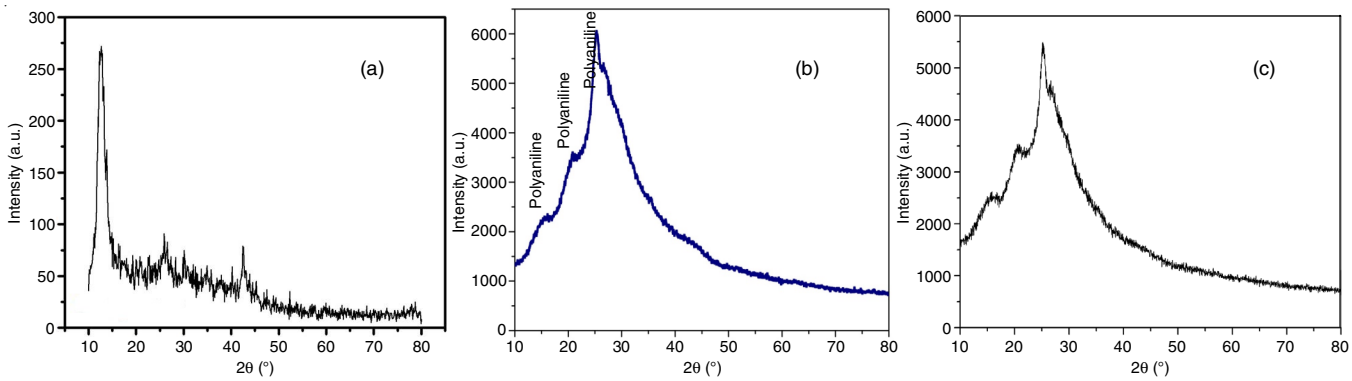



Fig. 2. XRD pattern of (a) GO nanocomposite, (b) polyaniline (PANI) and (c) PANI-GO nanocomposite 2 wt%

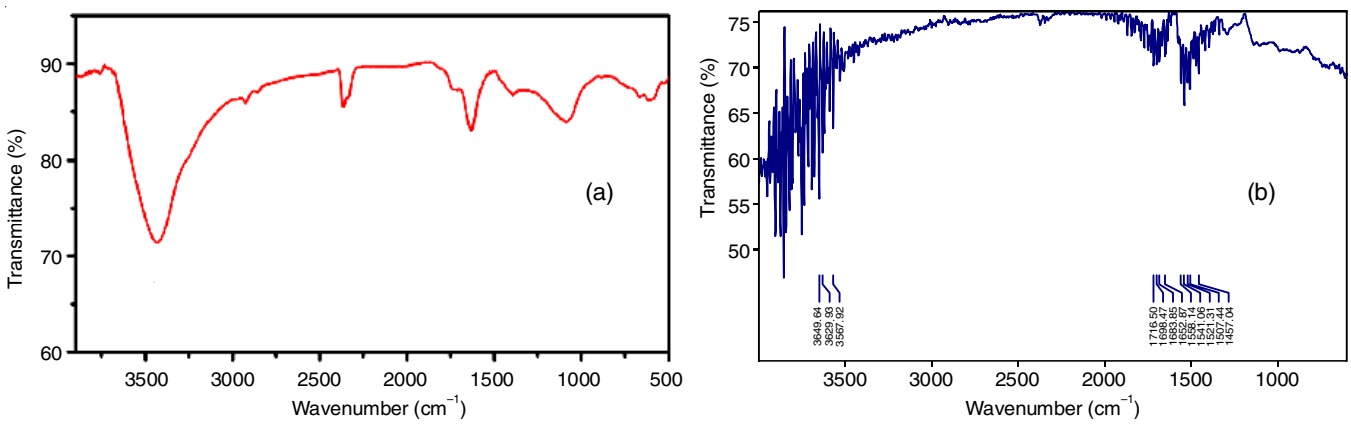



Fig. 3. FTIR spectra of (a) GO and (b) PANI/GO nanocomposite 2%wt

**FTIR studies:** The FTIR spectrum of graphene oxide is shows in Fig. 3. It shows a broad peak appeared at 3500 cm$^{-1}$ in the high frequency area whereas the FTIR spectra of PANI/GO are shown in Fig. 3b. The FTIR spectrum of PANI revealed the characteristics peaks of PANI, including the peak at 3567 cm$^{-1}$ that assigned to the N-H stretching. A peak at 1541 cm$^{-1}$ for the stretching vibration band of benzenoid moieties in PANI (C=C).

## Conclusion

A highly efficient and environmentally friendly method has been developed for synthesizing *bis*(indolyl)methanes. This method involves the electrophilic substitution reaction of 5-bromoindole with a substituted aromatic aldehyde in ethanol solvent utilizing a heterogeneous polyaniline supported graphene oxide catalyst. This approach aligns with the principles of green chemistry by avoiding the use of toxic catalysts. The proposed methodology offers numerous benefits, including environmentally benign conditions and a straightforward procedure. When compared to traditional approaches, this protocol has several advantages, including a shorter reaction time, mild reaction conditions and a reusable catalyst.

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## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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