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## Improvement on mechanical and flame retardancy behaviour of Bio- Exfoliated graphene-filled epoxy/ glass fibre composites using Compression moulding approach --Manuscript Draft--

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# Improvement on mechanical and flame retardancy behaviour of Bio- Exfoliated graphene-filled epoxy/ glass fibre composites using Compression moulding approach

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## Abstract:

Bio-reduction of composite materials is the modern approach to facilitate the researchers to avoid toxic chemical exposure during reduction process. In this study the green reduction of graphene oxide using *Abutilon indicum* (Tamil name: Thuthi) plant leaves, extracted from various solvents were used as green filler material (0.3, 0.6 & 1 wt.%) in glass fibre epoxy composite. Compression moulding process was adopted to fabricate bio-reduced graphene oxide (BGO) filler incorporated polymer matrix composite. Tensile, flexural, toughness, impact test and fracture surface morphology analysis have been conducted over developed composite. Flame retardancy behaviour was studied based on UL94 standard. Outcome of the study revealed that *Abutilon indicum* act as the better reducing agent to reduce graphene oxide. XRD, FTIR results depicts the proper distortion of graphite flask. Addition of BGO in polymer matrix improvises the tensile, flexural and impact strength of matrix material up to 28%, 55% & 80%. Herein addition of 0.6 wt. % of BGO showcase better fracture toughness and flexural strength further increment in BGO show negative effect in strength. Occurrence of delamination failure was notified in fracture surface morphology.

**Keywords:** *Abutilon indicum*; BGO; Compression moulding; flame retardant

## Introduction:

In recent days, reduction in carbon based emissions and fuel economy are the two main targets for transport industries. This fact assists in more consideration on development of light weight material with better performance. Among polymer based materials, fibre reinforced polymer composite material particularly attains more attention owing to its low density, high specific strength with better stiffness comparable to conventionally used steel[1,2]. Due to this facts fibre based polymer composite are consider as advanced engineering materials has wider range of applications that includes automobile, defence, aerospace etc. Herein epoxy an thermosetting based polymers that exhibits better toughness, chemical stability; hence used as matrix materials for developing polymer matrix composite and further these polymers are of low cost comparing to other polymers such polyester, vinyl etc[3,4]. In polymer matrix composite system glass fibres are used as reinforcement to improve the mechanical behaviour of matrix (polymers) materials. These glass fibres have high tensile strength and low cost while compare to available carbon and aramid based fibres. Hence, it has been majorly used in the field of transport industries and also in construction industries for retrofitting based jobs and roofing applications. Conversely, these glass fibre based polymer matrix composites exhibits brittle nature due to the presence of cross linked monomers in matrix material. Also, glass fibre based composite has poor out of plane properties that obstructs their potential usage in several engineering applications[5,6]. The flexural behaviour and out of plane properties of composite materials are primarily directed by interfacial interaction among the fibres and matrix materials. Additionally, lower toughness of polymers results in interlaminar failure, such as delamination. Likewise, epoxy based polymer materials are flammable and have major capability to produce volume of smoke and toxic gases during combustion that results in preposterous fire threats. To overcome these facts, phosphorous based compounds were used to improve the flame retardant behaviour of epoxy resin. However, addition of these compounds reduces the mechanical and thermal stability of epoxy. Hence there is a need for research based on improving the flame retardant behaviour of glass fibre reinforced polymer composite without affecting its bulk properties such as tensile, flexural, toughness strength etc[6–8]. Consequently, in light of this issue, material scientist and researchers have focus on using micro or nano fillers to improve the basic and functional properties of fibre reinforced polymer matrix composite. Some researchers depict that addition of these filler materials will definitely improve the interfacial bonding between matrix and fibre that improves its

strength. Likewise occurrence of interfacial interaction results in better energy transformation during fracture conditions that improves the fracture toughness and reduces the delamination based failures. Therefore, proper selections of filler materials must be made to attain required properties of composite. Amongst the available filler materials carbonaceous based fillers viz. CNT and various form of graphene viz. graphene nano plates, graphene oxide have attained substantial consideration from researchers. They reveal that addition of CNT and graphene improve thermo-mechanical behaviour of composites due to its high surface area and high aspect ratio, further act as better medium for transferring applied load from matrix to fibres thus increase the material performance during testing conditions[9–11]. Graphene based material showcase better thermal conductivity that help in avoiding the coincident of overheating and reduce the chance of fire spreading due to its better heat transfer ability[12–14]. There are few researches in which graphene derivatives are used to improve the mechanical and thermal stability of composites. Graphene nanoplatelet (GNP) was used as filler by Nicholas T. Kamar and co-worker to develop glass fibre reinforced polymer matrix composite. Ply layup method was adapted by them to develop high strength composite with varying percentage of GNP (up to 1 wt. %) and investigate its mechanical behaviour. Investigation shows that addition of GNP improves the flexural strength of fabricated composite up to a particular limit viz. 0.25 wt. % and further increment in filler addition exhibits reverse effect on its mechanical performance[15]. Hand layup method was utilized by Rajesh Kumar Prusty and co-worker to develop graphene oxide incorporated epoxy-glass fibre composite and investigate its flexural behaviour. Observations depict that addition of graphene oxide (upto 0.5 wt. %) improve the flexural strength of matrix material up to ~21%[16]. Kamila K et al developed GNP( upto 4 wt.%) reinforced polymer matrix by vacuum-assisted resin transfer moulding and examine the electrical and mechanical behaviour of composite. Outcomes expose that incremental percentage of GNP improve the surface and volumetric conductivity of developed composite and lower concentration of GNP displays better mechanical strength[17]. Solution blending method was adopted by Shan Liu et al to develop Graphene Nanosheet (GNS) filler incorporated epoxy matrix composite and investigate the synergetic effect of GNS over flame retardant behaviour. Observation depicts that addition of GNS improve the flame retardancy of matrix material and increase the limiting oxygen index of epoxy[18]. Bin Y et al had found that usage of phosphorous and nitrogen based functional graphene oxide in epoxy matrix increases the flame retardant performance of epoxy resin[19].

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Currently, the theme of green chemistry has been accentuated in educational societies since it can effort on developing newer protocol for chemical products by eradicating the cohort of risky substances. With the progress of nanotechnology, the philosophies of green chemistry have extensively practised in synthesis and usage of nanomaterials. In green synthetic approach material synthesis has, two features that includes consumption of nontoxic chemicals and eco-friendly solvents have concerned significant responsiveness owing to their benefit in reducing the environmental risk. In earlier graphene was synthesis by CVD method, conversely it have some drawbacks like high cost and poor repeatability. Further modified Hummer's are used for exfoliation of graphene form graphite flakes whatsoever to reduce the synthesis graphene oxide high toxic chemical such as hydrazine hydride, hydroquinone are used and the by products from reduction process are not eco-friendly and may cause serious effect in human societies. In order to overcome this fact, natural products and micro-organism viz bacteria are used for reducing graphene oxide by replacing the toxic reducing agent. Green plant extract contain natural anti-oxidants such as eugenol, anthraquinone acetylated mannans ascorbic acid, anthrones polyphenols having better efficient to reduce graphene oxide[20–22]. Suhasini et al. used *Ocimum sanctum* (Tulasi leaf) extract to reduce graphene oxide and attain better reduction in synthesising reduce graphene oxide. In some studies wild carrot and Aloe vera extracts were used as reducing agent and the reduce graphene oxide exhibit better performance in photo catalyst and electrochemical applications[23]. *Abutilon indicum* (Tamil: Thuthi), a medicinal plant, which are abundantly available in southern region of India. It is known for its medicinal value, which is used in Siddha, Ayurveda and Unani medicine the treatment of gastric disorder, arthritis, cancer, neuro degeneration and hepatic disorder. The extract of *Abutilon indicum* leaves consist of natural antioxidant alkaloids, phenols, steroids and terpenoid, which are the natural source of active reducing agent[24].

Based on the clear cut literature survey it can be depicted that additions of graphene improve the basic and functional behaviour of epoxy while used it as filler material. Additions of lower concentration of graphene increase the thermo-mechanical behaviour of glass fibre composite. Usage of plant extract helps in effective reduction of graphene and promoted eco-friendly effects. *Abutilon indicum* have natural antioxidant with high medicinal values, however usage of *Abutilon indicum* as reducing agent was not yet reported. Based on this fact, an attempt have been made to reduce, graphene oxide by *Abutilon indicum* leaf

extract and using the Bio-reduced Graphene oxide (BGO) as filler material to improve the mechanical and flame retardancy behaviour of epoxy- glass fibre composite.

## 2. Materials and method:

### 2.1. Materials:

Epoxy resin diglycidyl ether of bisphenol A grade LY 556 along with HY951 grade ( $C_6H_{18}N_4$ ) hardener possessing 2 to 3 h gel time in general was used as the matrix. Utilization of the said material is recommended as because of its lower viscosity range. Glass fibre with 200 GSM was used as reinforcement to improve the mechanical strength of epoxy polymer. Recent studies revealed that addition of glass fibre up to 30 wt. % exhibits better mechanical strength hence 30 % of glass fibre was kept constant to developed novel composite. Graphene oxide was synthesis by modified hummer's method [25]. *Abutilon indicium* leaf extract was used to reduce the graphene oxide to avoid the usage of harmful chemicals such as hydrazine hydride, which cause several health issues. Further, the *Abutilon indicium* plant extract have several natural anti-oxidants properties with the presences of alkaloids, flavonoids, phenolic compounds, tamins, saponins, sterols. Similarly, addition of graphene fillers up to 1 wt. % improves basic and functional properties of matrix material. Therefore, composites were developed with varying percentage of bio-reduced graphene fillers (0.3, 0.6 & 1 wt.%).

### 2.2 Biosynthesis of filler material:

#### 2.2.1 Preparation of Bio-extract:

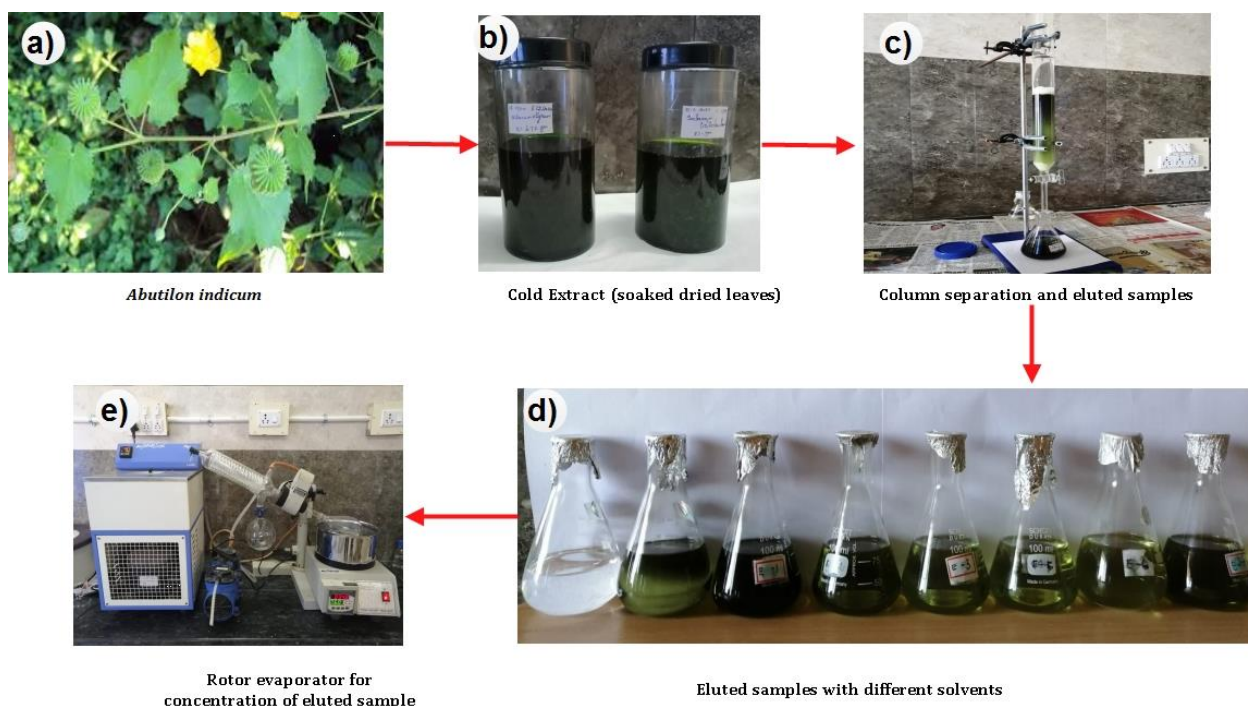
The *Abutilon Indicum*, a presumed traditional medicinal plant (Figure 1.a) was used in this study for its effective antioxidant properties. The leaves were collected from Karaikudi, Sivagangai District, Tamilnadu and identified by Dr. C. Rajasekar, Dept. of Botany, Alagappa University, Karaikudi, Tamilnadu, where a voucher specimen has been preserved for future identification. The collected leaves were washed with distilled water and shade dried for 3-5 days and was broken into a coarse material. About 500 gm of the coarse material was subjected to "Cold Maceration"(Figure 1.b) for 15 days, soaking in low polar solvents, ethanol and double distilled water in the ratio of 3:1 with a periodic stirring.

After maceration, the solvent was then filtered using whatman filter paper and the filtrate was concentrated to the consistency of semisolid crude extract using a hot air oven at

temperature ranging 37-40°C. The semisolid crude extract was then subjected to the preparation of slurry by gently mixing with silica gel (60-120) at the ratio of 1:3. The slurry was then stored in airtight container at room temperature. About 5 gm of slurry sample was eluted with various solvents viz petroleum ether, chloroform, ethyl acetate, ethanol and water. The eluted extracts from various solvents were collected individually (Figure 1.d) and concentrated using rotar-evaporator (Figure 1.e). The presences of active compounds were confirmed by TLC profiling.

### 2.2.2 Thin layer chromatography profiling of various extracts:

The TLC profiling was performed as described by Biradar et al., 2013[26]. Various solvent extracts of *Abutilon Indicum* leaves were subjected to TLC to validate the presence of active compounds. The TLC plates were prepared by using Silica gel 'G' as 30 gm of silica gel was weighed and made to a homogenous suspension with 60 ml distilled water for two minutes, this suspension was distributed over the plate, which was air dried until the transparency of the layer disappeared. The plates were dried in hot air oven at 110° C for 30 mins and then stored in a dry atmosphere and used whenever required. Samples were prepared by diluting the crude extracts of chloroform, acetone, ethanol and water with respective solvent and then applied usually 1-10 µl volumes to the origins of a TLC plate 2 cm above its bottom with the help of capillary tubes.





**Figure1 (a-e) Process outline of *Abutilon Indicum* leaves extraction process.**

### **2.2.3 Synthesis of *Abutilon indicum* derived graphene oxide:**

Graphene oxide was synthesis by modified hummers method, the detailed methodology was depicted in our previous reports [14]. Green reduction of graphene oxide was carried out using *Abutilon indicum* plant leaf extract dissolved in ethanol. The attained graphene oxide was dispersed in distilled water in ratio of 1mg/ml. Herein, 50 mg of GO was dispersed in 50 ml of distilled water and ultrasonicated for 1 h it is followed by addition of 50 ml of green extract. The mixer was ultrasonicated for 1 h to attained uniform dispersion and further heated at 90° C with continues stirring for 12 h. The obtained mixture was washed with distilled water and ethanol using cooling centrifuge rotates at 10,000 rpm. The achieved sample was vacuum dried for 24 h and named as Bio-reduced Graphene Oxide (BGO).

### **2.3 Composite fabrication:**

Compression moulding method was adapted to developed BGO based polymer matrix composite. Herein, calculated weight percentage of BGO was dispersed in ethanol and ultrasonicated for 1 h after that the dispersed BGO fillers are added into epoxy resin. The matrix and filler materials are mechanically stirred for around 1200 rpm for 2 h to attain uniformly dispersion of BGO filler in matrix materials. After this HY951 hardener was added in 1:10 ratio and mixed for five minutes. Formerly the attain resin mixtures are coated over both side of glass fibre using hand layup method. 10 layers of resin/fillers coated glass fibre are stacked in flat mould of dimension 270 x270x30 mm and degassed in vacuum bucket. After degasification process the stacked material was hot pressed for around 15 MPa and allowed to cure, the attain samples were names as BGOX-GF-Epoxy composite. The same procedure was followed to develop BGO based composite herein X denoted the percentage of BGO filer wt. % viz. 0, 0.3, 0.6, 1 wt. %.

### **3. Material characterization:**

X-ray diffractometer (BRUKER) was utilized to endorse the presence of reduction of green synthesised graphene oxide. The synthesised graphene oxide was scanned at 2 theta ranges



from 20° to 60° and the attained patterns are matches with JCPDS card for phase confirmations. Spectrum GX, Perkin Elmer, Fourier Transform Infrared spectroscopy (FTIR) was used to confirm the presence of functional group in plant extract and BGO sheets. TLC study was to validate the presences of active compounds in the bio-extract. ASTM D 638 standard was adopted to understand the tensile behaviour of developed composite. ASTM D 790 and ASTM D 256 standard are used to calibrate the flexural and impact strength of fabricated composite. Fracture toughness of composites was measure based on ASTM D5528 standard. Experiments were repeated for four times and the average value was noted for exactness of the attained results. Scanning electron microscopy (JEOL JSM6610LV) was used for fracture surface analysis. UL-94 vertical burning test was accompanied by adopting ASTM D3801 standard and tests were conducted over five samples and the average values are noted. Limiting oxygen test was conducted on developed samples based on ASTM D2863.

## 4. Results and discussion

### 4.1 Confirmation test for plant extract:

#### 4.1.1 Thin Layer Chromatography of plant extract:

The TLC results showed the presence of active compounds in chloroform, ethyl acetate and ethanol extracts. Each band showed in the picture confirms the presence of active compounds. The chloroform extract showed the presence of high number of active compounds than that of ethyl acetate and ethanolic extract.

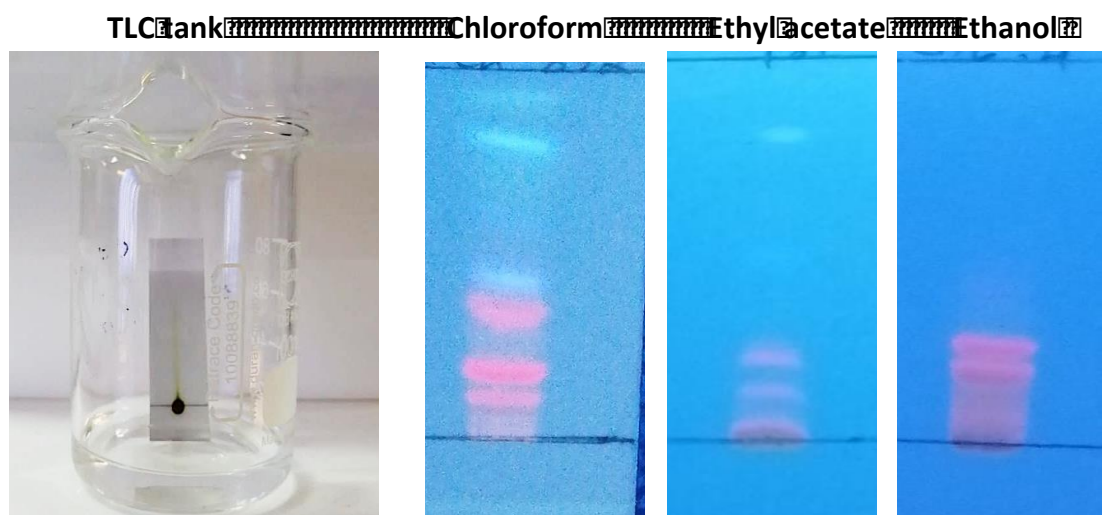
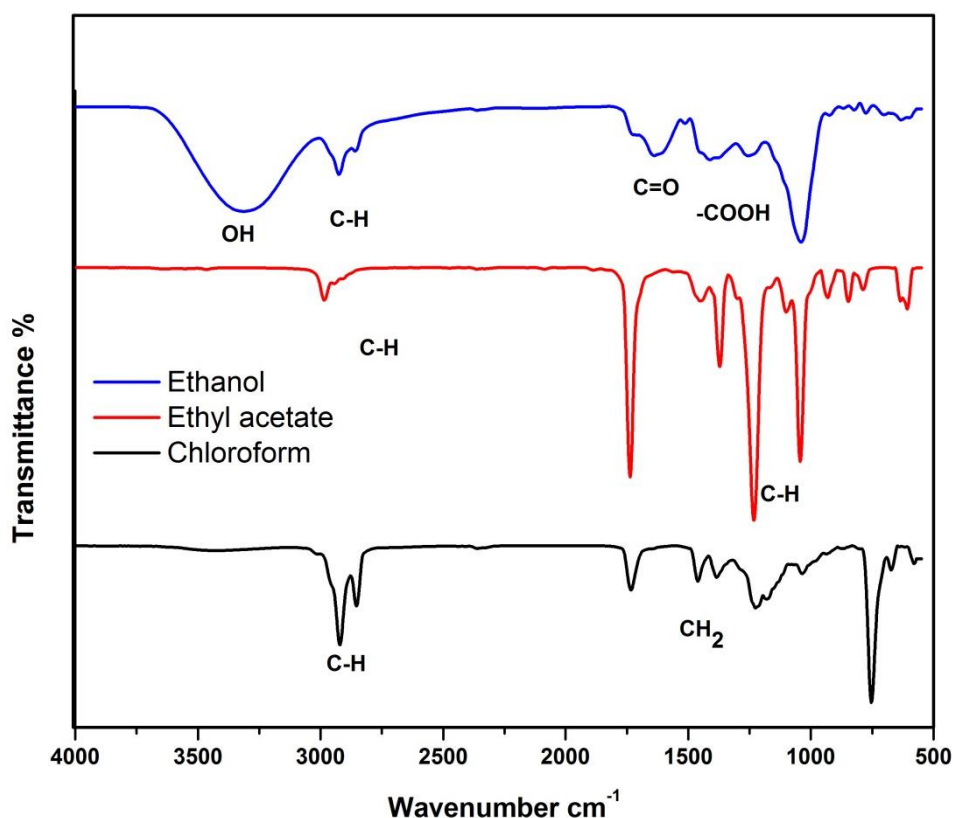


Figure 2 Thin Layer Chromatography of *Abutilon Indicum* leaves extract

#### 4.1.2 FTIR Analysis of *Abutilon Indicum* leaves extract:

Functional groups confirmation was carried out with the help of FTIR, figure 3 depicts the FTIR results of *Abutilon indicum* extract dissolved in organic solvents like ethanol, ethyl acetate and chloroform. Presences of phenol group with OH stretching are noticed at  $3300\text{ cm}^{-1}$  in ethanol extract. The broad peaks ranges from  $3000\text{ cm}^{-1}$  to  $2500\text{ cm}^{-1}$  are related to carboxylic acid and alkanes group with C-H stretching vibration, the peaks with range of  $1900\text{ cm}^{-1}$  to  $1500\text{ cm}^{-1}$  are attributes to amide based functional group. Minor peaks at the range of  $2200\text{ cm}^{-1}$  to  $2300\text{ cm}^{-1}$  confirms the presence of Nitrile group  $\text{C}\equiv\text{N}$ . Herein narrow peaks near  $1590\text{ cm}^{-1}$  are attributes of C=O stretching of amide based functional groups; peaks near  $1410\text{ cm}^{-1}$  are related prevalence of alkanes groups with COOH and  $\text{CH}_2$  bending vibration. Sharp narrow peaks near  $1380\text{ cm}^{-1}$  is also related C-H stretching of alkanes group[27].

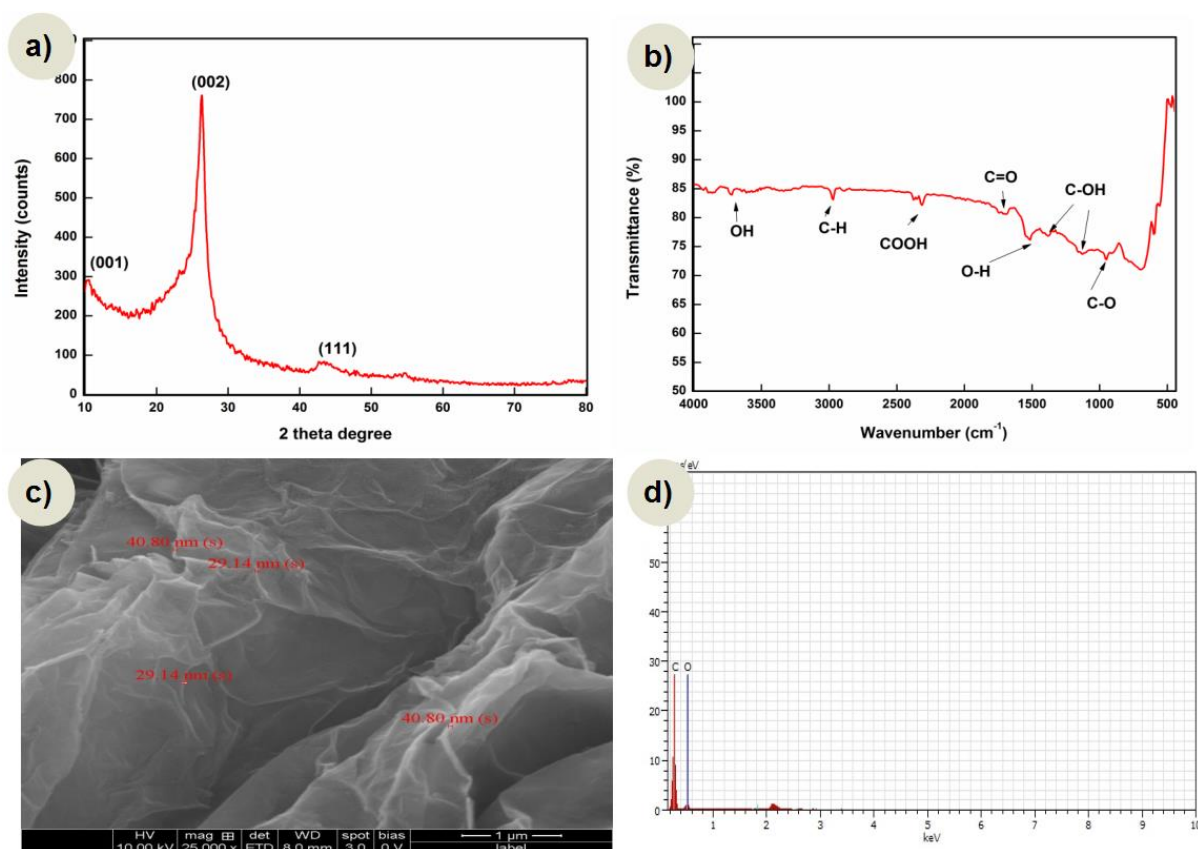


**Figure 3 FTIR of *Abutilon indicum* extract under various organic solvent**

Existences of skeletal vibration for C-H stretching were notified at the range of  $1100\text{ cm}^{-1}$  to  $1200\text{ cm}^{-1}$  has belonged to presence of isopropyl group. Slight peaks are identified in-between  $700\text{ cm}^{-1}$  to  $550\text{ cm}^{-1}$  that confirms the presences of alkenes group with C-H plane banding. Peaks at the range of  $1100\text{ cm}^{-1}$  to  $1250\text{ cm}^{-1}$  are conforming to the presence of carboxylic acid, ether and alcohol with O-H functional groups. Peaks near  $1040\text{ cm}^{-1}$  related to the

presence of sulphur based amino acids[28,29]. The FTIR spectrum confirmed the presence of phenols, amino acids, amides, carboxylic acids, alkanes, aliphatic esters, secondary alcohols, sulphur compounds and mono substituted alkenes in leaf extract of *Abutilon indicum*. Further the presences of functional groups are traced more in ethanol extract and hence *Abutilon indicum* extract dissolved in ethanol is chosen for further reduction process of graphene oxide.

#### 4.2 Confirmation test for bio reduced graphene oxide



**Figure 4. Bio-reduced graphene oxide a) XRD b) FTIR C)SEM D) EDAX**

The confirmation test for green synthesized graphene oxide was depicted in figure 4(a-d). Herein XRD of BGO was depicted in figure 4a. The peak near 10.3° was associated with graphene oxide with (001) plane. The peak near 25.4° reveals the reduction of graphite. In other word, oxidation of graphite to graphene oxide reflected in the occurrence of peak ( $\theta=25.4^\circ$ ) with (002) plane[30]. In this case graphitic peak shift toward 25.4° this might due to oxidation of graphite. The presences of function group and antioxidants in the green extracts might be the reason for oxidation of graphite.

Further many minor peaks are notified from XRD graph this might be due to presence of disorder or unreduced graphite and presence of phenolic groups. Additional peaks near  $42.1^\circ$  represent (111) plane and reveal the reduction of graphene oxide[31,32]. Minor peaks are notified at the range of  $50^\circ$  to  $55^\circ$  that might the (004) plane of graphite. FTIR results of synthesised BGO were depicted in figure 4b that help in identifying the modes corresponding oxygen containing functional group. Herein peaks at the range of  $3800\text{ cm}^{-1}$  to  $3300\text{ cm}^{-1}$  are related to the stretching vibration of OH group. The peak arises near  $2854\text{ cm}^{-1}$  is related to C-H stretching vibration. These hydroxyl groups might absorb water molecules and phenolic functional group from the carboxylic group of green extract[25]. C=O stretching vibration of carboxyl groups was detected at  $1672\text{ cm}^{-1}$ . The presence of carboxylic group help in increasing the interaction between the polymer materials that helps in better adhesion. The peaks near  $\sim 1750\text{ cm}^{-1}$  may be due the occurrence of stretching vibration of C=O and further indicate the reduction of graphene oxide. Peak near  $1431\text{ cm}^{-1}$  indicate the presence of O-H group with deformation vibration. Peak near  $1300\text{ cm}^{-1}$  and  $970\text{ cm}^{-1}$  are related to the presence of C-OH stretching vibration group and C-O functional group[33,34]. SEM and EDAX of BGO were depicted in figure 4(c-d), the average size of graphene oxide sheet was measure as  $\sim 50\text{ nm}$ . And the EDAX results show the presence of carbon and oxygen elements in BGO.

#### 4.3 Mechanical behaviour of developed composite

##### 4.3.1 Tensile strength and percentage of elongation

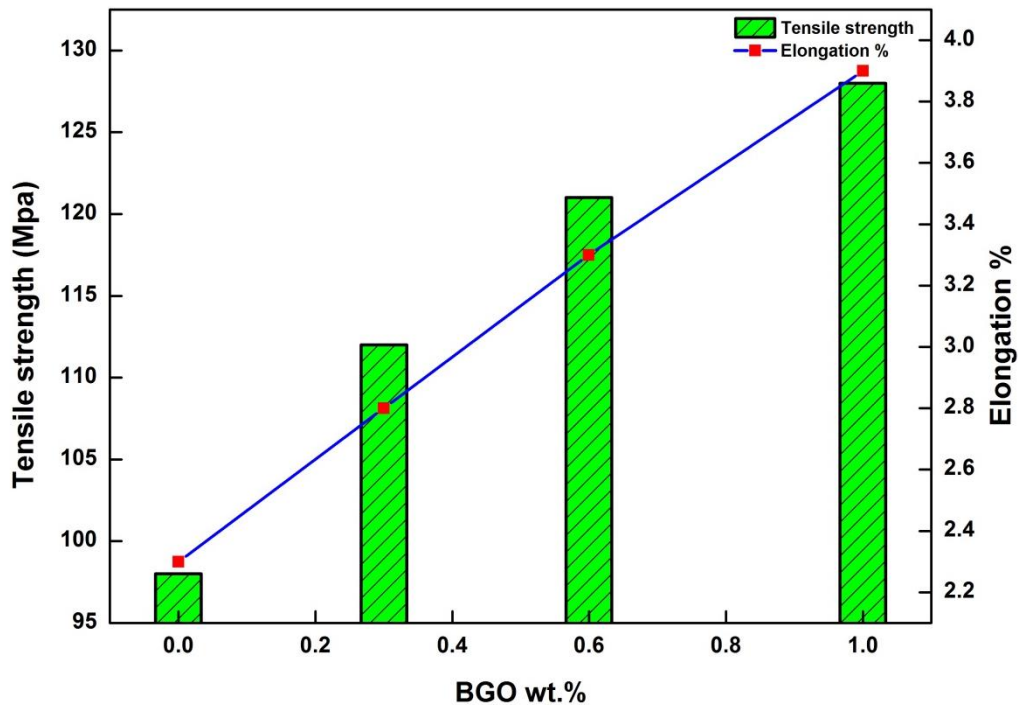


Figure 5 Variation of Tensile Strength with respect BGO addition.

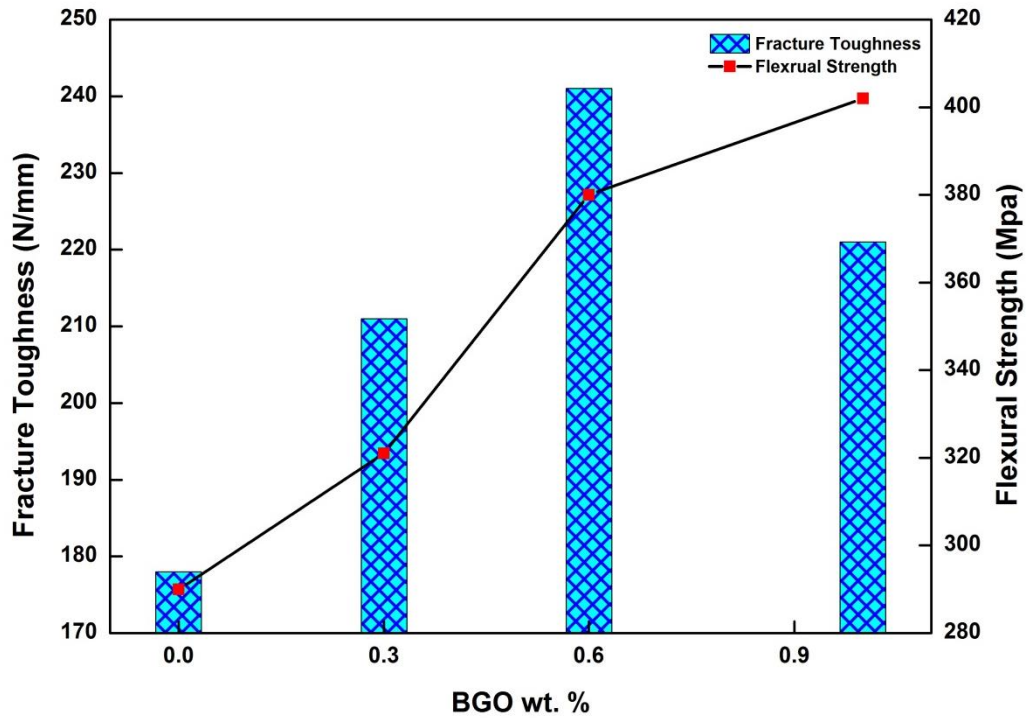
Figure 5 depicts the influence of BGO on tensile behaviour of developed composite. It can be notified that addition of BGO improves the tensile strength of composite. This might be attributes to the biological reduction of graphene oxide. Addition of BGO improves the adhesion between the matrix and glass fibre that improves the tensile strength. Herein better tensile strength of matrix materials are mainly depends on aptitude of matrix or polymer to transfer the applied load to the refinements viz. glass fibre, addition of BGO based filler have better interface strength with matrix materials that helps in effective transfer of applied stress over the reinforcement phase of composite material. Conversely the interfacial bonding between the filler and matrix material are mainly depends on mechanical and chemical interaction that influence the molecular nature of both component in matrix and reinforcement phases[14]. The interfacial region between the filler and matrix materials acts as the major parameters in improving the strength of developed composite with varying addition of BGO. BGO based fillers exhibits better surface area that help in improving the interfacial bonding even at low wt. % of BGO. Hence, the stress transfers from matrix to filler that permit the fabricated composite to withstand higher stress. Inspect of better interfacial bonding, these developed composite have high strength compared to matrix material owing to physisorption of matrix over the surface of filler material[15]. It can also be infer that addition BGO increase the plastic deformation of epoxy composite that results in higher elongation as depicted in figure 5 while compare to matrix material. Presences of BGO delay the duration on crack linking initiation and de-bonding in polymer matrix that ultimately increases the tensile strength of the developed composite[16,17]. Comparisons of tensile strength with previous literatures are depicted in table 1.

**Table 1 Comparison of tensile strength with available literatures**

Matrix	Filler	Tensile strength (Mpa)	Reference
Epoxy+ Glass fiber	Nano graphene = 1, 2 %	Ts = ~78,~168	[35]
Epoxy+ Glass fiber	BGO = 0.3,0.6,1	Ts = ±112,±121,±128	Current research

#### 4.3.2 Flexural and toughness behaviour of composite





**Figure 6 Variation of fracture toughness and flexural Strength with respect BGO addition**

It can be notified from figure 6 that addition of BGO up to 0.6 wt. % shows better toughness and further increment in percentage decreases the toughness nature of composites. However flexural strength of the composites increases with respect to the addition of BGO nano fillers. The synthesis BGO filler have various functional groups such as carboxylic that helps in improvising the interfacial bonding between the matrix and reinforcement phase that increase the stress distribution ability of matrix material that's results in improved mechanical strength .Addition of BGO increases the hydrogen bonding between the filler and matrix that might be the reason for decreased strength. Addition of BGO enhance the interfacial interactions between the glass fibre and matrix, this could effectively improve the load transfer efficiency that lead to higher flexural strength. The BGO filler has high surface area hence increase in BGO content initiates the agglomeration in matrix phase that decrease the net free energy hence lower fracture toughness was notify beyond 0.6 wt. % of BGO. Increase in BGO increases the clustering effects that decrease the aspect ratios in composite phase[36,37]. These facts results in decrease in interfacial region among the matrix and reinforcement and further reduce the stress transformation tendency of matrix material that decrease the composite toughness. However, increase in addition BGO show positive trends in flexural properties; presences of BGO filler results in crack deflection and crack branching

1 in composite phase owing to its higher energy absorption capacity and high surface area thus  
2 increase the strength. Addition of BGO initiates inters lock effect between matrix and fibre  
3 phase due to the it's wrinkled morphology that improves the composite strength. Further,  
4 presences of BGO increase the stability of composite phase by initiating better adhesion  
5 between epoxy and glass fibre. Weight percentage of BGO has two effect in failure mode,  
6 herein material failure occurs due to the lower wt. % of BGO due to the tension side and  
7 increase in BGO wt.% results in failure on compression side[38].  
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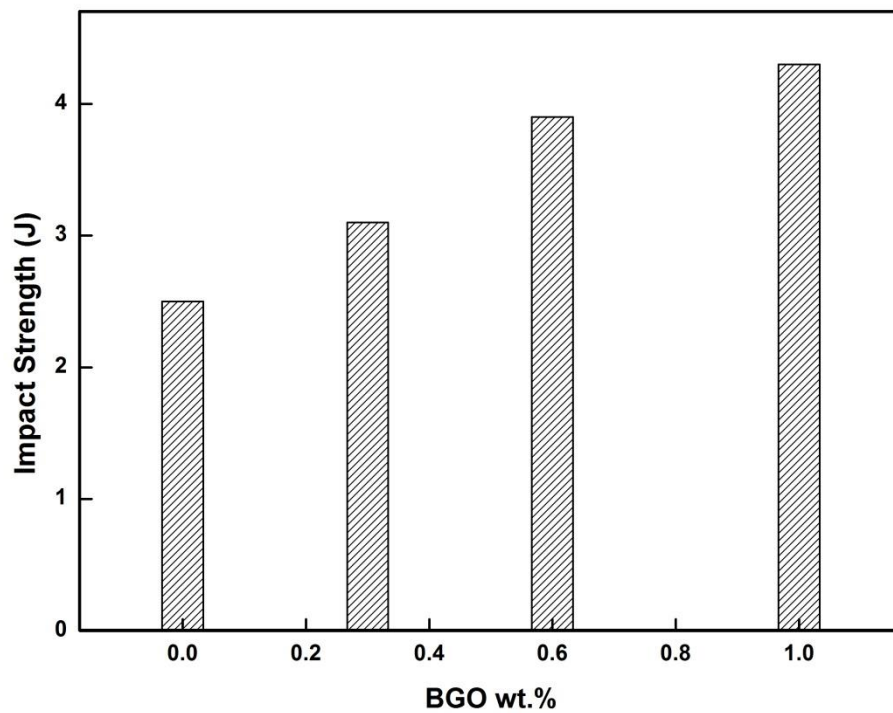
13 Herein, lower BGO wt. % increase the stress transfer efficiency among the  
14 polymer and reinforcements, conversely increase in BGO addition decrease the inter laminar  
15 adhesion that results in micro buckling and delamination failure thus reduce its strength.  
16 Addition of BGO beyond 0.6 wt. % decreases the fracture toughness due to clustering effects  
17 and these cluster acts as stress connectors and pave the way for crack propagation by forming  
18 sub micro voids. Herein void formation would lead to weaker interlaminar strength and  
19 inadequate adhesion among the matrix and reinforcement thus decrease its strength[39].  
20 Further, cracks initiate and grow from the void coalescence and agglomerates, with continued  
21 loading, fibre breakage, matrix embrittlement, de-bonding, and delamination could occur,  
22 resulting in catastrophic failure among the fibre matrix composite structures. On another view  
23 crack tip bifurcation upon its meeting with filler, separation between the graphitic layers and  
24 shear failure of matrix are the extra failure mechanisms. Furthermore, the strong p-p  
25 interactions between filler contribute to the stacking of individual sheets. Some mechanism  
26 for better fracture toughness may be shear yielding, crack deflection, and crack pinning and  
27 bowing. Additions of BGO filler transfer the brittle nature of epoxy and increase the ductile  
28 properties thus increase the mechanical strength[40–42]. This might be due to the surface  
29 roughness of BGO. The matrix material have low surface roughness thus results in brittle  
30 fractures. Comparisons of flexural strength with previous literatures are depicted in table 2.  
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**Table 2 Comparison of flexural strength with available literatures**

Matrix	Filler	Flexural strength (Mpa)	Reference
Epoxy+ Glass fiber	Nano graphene = 1, 2 %	Fs = ~375,~527	[35]
Epoxy+ Glass fiber	GO = 0.1, 0.5, 1 %	Fs = ~378, ~393, ~364	[43]
Epoxy+ Glass fiber	2Si + GO= 0.1,0.5 %	Fs = ~369, ~ 358	[44]
Epoxy+ Glass fiber	BGO = 0.3,0.6,1	Fs = ±321, ±380, ±402	Current research

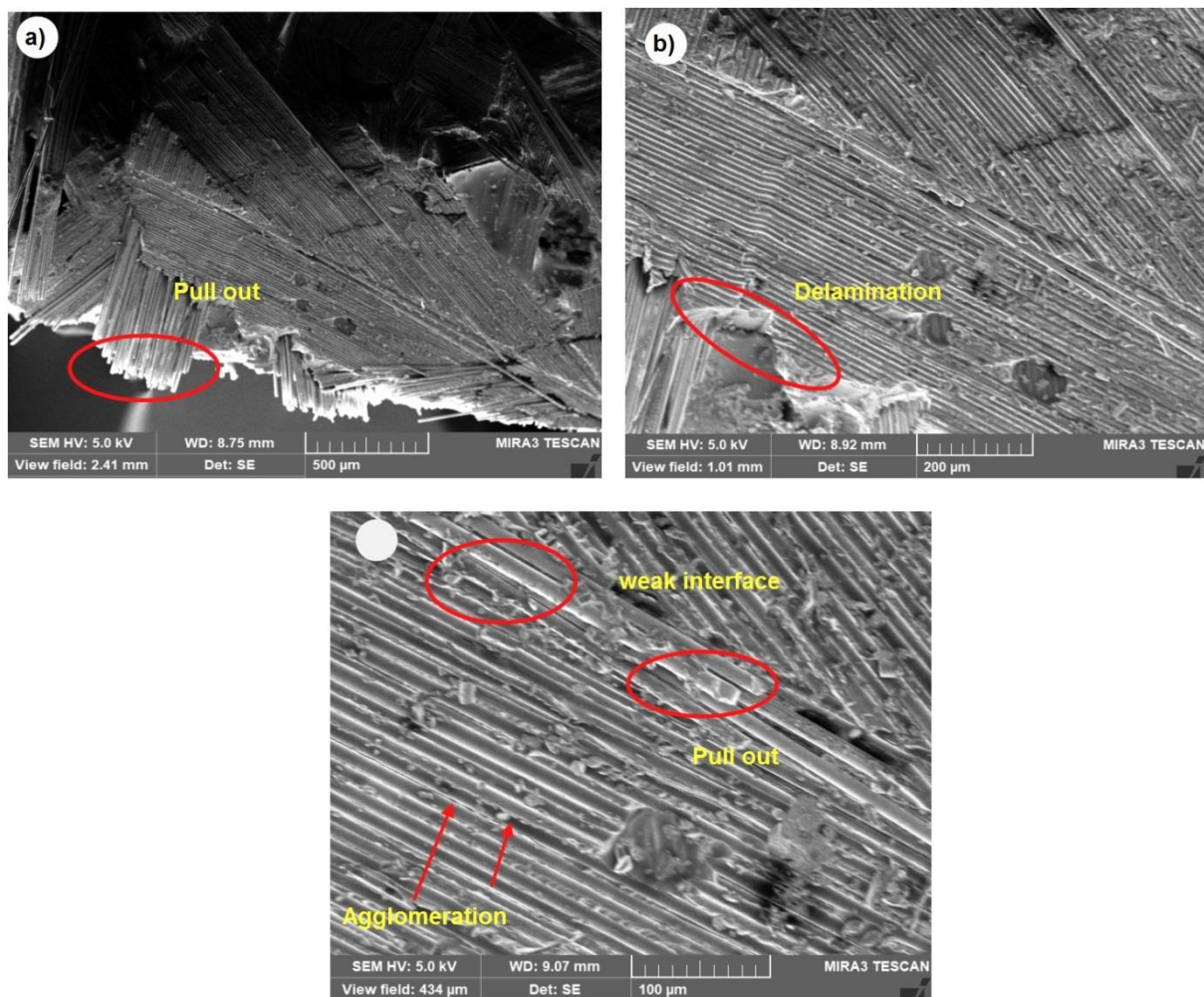
#### 4.3.2 Impact strength of composite



**Figure 7 Variation of impact Strength with respect BGO addition**

Figure 7 depict the influence of BGO on impact strength of the developed composite. It can be notified that incorporation of BGO in epoxy matrix improves the impact strength of develop composite. These BGO filler have high surface area and have better energy absorption capacity, this fact results in improved impact strength. BGO filler uniformly dispersed over the matrix phase and form well inter lock between the matrix and glass fibre. That assists in absorbing the impact energy thus improves the strength. Presences of BGO filler results in crack deflection and crack branching in composite phase owing to its higher energy absorption capacity and high surface area thus increase the strength[45]. These winkled shape nano filler uniformly dispersed over matrix material that deflect the crack growth that increase its impact strength. As a result, the crack is forced to move out of its initial propagation plane by tilting and/or twisting. BGO based filler materials increase the adhesion bonding in composite phase that increase the resistance of developed composite against bending thus exhibiting better ability to absorb energy near the impact regions[14,46].

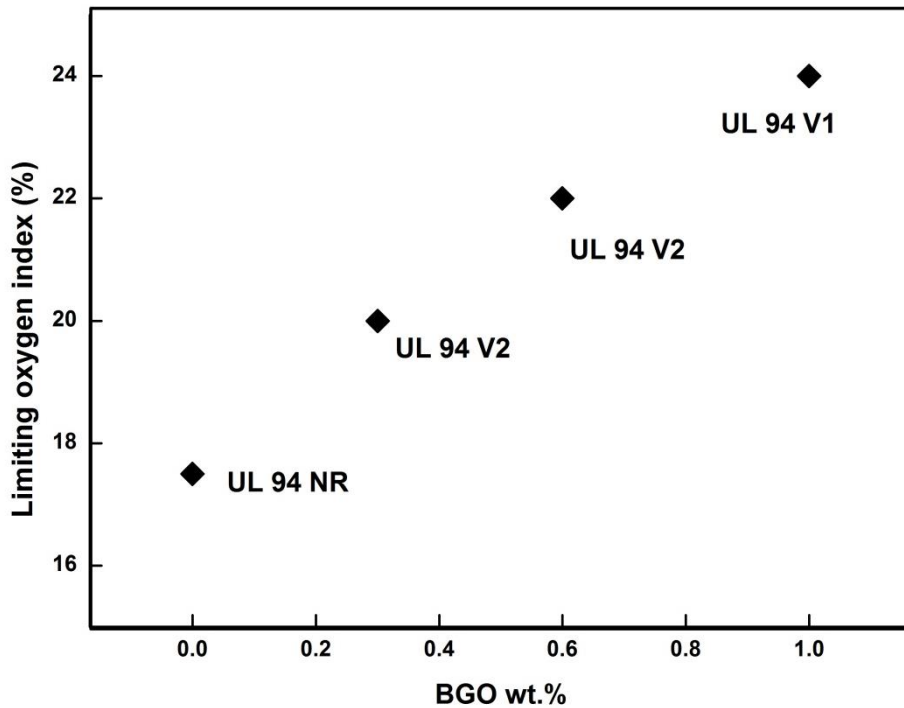
#### 4.3.4 Fracture surface morphology of composite



**Figure 8 Fracture surface of composite with: a) 0.3 wt. % of BGO; b) 0.6 wt.% of BGO  
c) 1 wt.% of BGO**

Figure 8 depicts the fracture surface of the developed composite with varying addition of BGO. Formation of pull outs are notified in figure 8a it can be related with the amount of epoxy attached on pulled out fibres owing to its better interfacial adhesion bonding among the matrix and reinforcement phase. The interactions between the epoxy and reinforcement delivers proficient stress transformation from the polymer to matrix phase that decrease the chance of crack progression rate, ensue in better mechanical strength of fabricated composites. Delamination and deformations are evident in composite phases that might be due to the absorption of applied energy during testing condition (figure 8b). Fracture failure occurs mostly within the matrix nearby the glass fibre surface because of matrix cracking and its deformation. It can also be justified that adhesion bonding between matrix and reinforcement phase are optimum up to 0.6 wt. % of BGO. Further increment in BGO results in weaker interfacial bonding as noticed in figure 8c. Further it can be also be infer that lower interfacial bonding among the composite and refinement might be due to the agglomeration tendency of BGO that reduces the bonding between the polymer and reinforcement phase[39]. However in case of epoxy-glass fibre-BGO composite, addition of BGO increase the tensile strength of developed composite that might be attribute to high surface area of BGO and ability of GO that decrease the brittle nature of epoxy matrix thus increase the ductility that results in improved tensile strength. Presences of matrix material are inferred in fibres that indicate BGO has better stability to increase the interfacial adhesion bonding among the matrix and reinforcement phase. It is feasible that during crack propagation the synergistic effect of BGO act in order to deflect its smooth path. Primarily BGO prevents the possibility of growth major cracks and resist the propagation to smaller ones. Thus inculcation of BGO, results in improvement on interlaminar fracture energy of developed composite[37,46].

#### ***4.4 Flame retardant of developed composite***



**Figure 9 Limiting oxygen index and vertical flammability ranking of composites**

Limiting oxygen index and UL 94 rating are depicted in Figure 9. Herein UL 94 ratings are given based on the burning seconds. Based on available literatures burning seconds less than 10 are termed as V0 rating and the developed material is consider as the best for flame retardant applications. V1 ratings are given to the sample which has burning seconds of less than 30 seconds and not sustained. V2 rating is given to the samples with burning seconds of 30 and is not sustained, but flaming drips ignite. NR rating is given to the samples which have more than 30 seconds of burning. Limiting oxygen index value is an important pointer on flammability of the polymer and plastic materials as in general, materials with higher limiting oxygen index are consider as the flame retardant material. Limiting oxygen index is defined as the volume percentage of minimum oxygen consideration that assists in materials combustion[47,48]. It can be notified from figure 9 that addition of BGO show case better influence in limiting oxygen index of epoxy polymer matrix composite. Incorporation of BGO in epoxy matrix improves the limiting oxygen index values up to ~37.1 %. In this addition of 0.6 wt.% of BGO improves the limiting oxygen index values up to ~ 25.7 % and ~14.2 % of improved limiting oxygen index values was attained for 0.3 wt.% of BGO filler. It can also be notified that matrix material have poor limiting oxygen index, which indicates the poor flammability resistance of epoxy

polymer[49]. It can also be observed that addition of BGO show case excellent performance on flame retardant behaviour of developed composite. This might be due to the effect of intermolecular and intramolecular interaction among the matrix and filler phase. During testing conditions, presence of BGO fillers helps in limiting the rapid burning of composite in thermal condensation of functional groups present in composite structures. Presence of BGO filler forms as passive barrier over the burning surfaces that hinders the proclamation of non-combustible gases and further limits the destruction of dripping properties of composite. It can be notified that increase in addition of BGO reduces the burning timing of developed composite[18]. The samples with higher BGO wt.% has V1 rating in vertical flammability testing, it depicts that increasing in BGO improves the flame retardant performance of developed composite. V2 rating are attained for composite made up of 0.3 and 0.6 wt. % of BGO, this might be due the insufficient supply of filler material that reduce the burning process. The matrix material have NR rating that means the samples have burning timing more than 30 seconds or burned completely.

## Conclusion:

Bio-reduced graphene was synthesized by *Abutilon indicum* leaf extract, which is used as filler material to improve the mechanical and flame retardancy of epoxy matrix. The present study on bio-reduced graphene revealed the potential reduction property of *Abutilon indicum* leaf extract, which enlightened the novel pathway for its use in field of composite synthesis. Addition of BGO results in better improvement in mechanical properties viz. 28% improvement in tensile and 80 % impact behaviour of composite. It has been observed that incorporation of 0.6 wt. % BGO improves the fracture toughness of composite up to 36% and 55% of flexural strength. Flame retardancy studies revealed that V1 rating was attained for 1 wt. % BGO incorporated in matrix material and decrease in BGO wt. % reduce UL 94 rating. The developed composite better mechanical and stability to flame thus it might be suggested for potential applications such as roofing sheet and car dash boards.

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## Response to reviewer:

### Reviewer #1:

*Comment 1: The abstract part needs to include mathematical findings to be more informative.*

*Response: As per reviewer suggestion mathematical finding has been updated in this revised manuscript*

Addition of BGO in polymer matrix improvise the tensile, flexural and impact strength of matrix material up to 28%, 55% & 80%.

*Comment 2: The introduction needs to put forward the highlights of your research. Please follow the literature review and show the knowledge gaps identified and link them to your research objectives*

*Response: Clarifications has been made as per reviewer console*

Based on the clear cut literature survey it can be depicted that additions of graphene improve the basic and functional behaviour of epoxy while used it as filler material. Additions of lower concentration of graphene increase the thermo-mechanical behaviour of glass fibre composite. Usage of plant extract helps in effective reduction of graphene and promoted eco-friendly effects. *Abutilon indicum* have natural antioxidant with high medicinal values, however usage of *Abutilon indicum* as reducing agent was not yet reported. Based on this fact, an attempt have been made to reduce, graphene oxide by *Abutilon indicum* leaf extract and using the Bio-reduced Graphene oxide (BGO) as filler material to improve the mechanical and flame retardancy behaviour of epoxy- glass fibre composite.

*Comment 3. The authors should be noted that comparing current results with published data can strengthen the impact of the study. Please add the comparison in the revision.*

*Response: As per reviewer advice comparison tables has been added in this updated manuscript*

**Table 1 Comparison of tensile strength with available literatures**

Matrix	Filler	Tensile strength (Mpa)	Reference

Epoxy+ Glass fiber	Nano graphene = 1, 2 %	Ts = ~78,~168	[37]
Epoxy+ Glass fiber	BGO = 0.3,0.6,1	Ts = ±112,±121,±128	Current research

**Table 2 Comparison of flexural strength with available literatures**

Matrix	Filler	Flexural strength (Mpa)	Reference
Epoxy+ Glass fiber	Nano graphene = 1, 2 %	Fs = ~375,~527	[37]
Epoxy+ Glass fiber	GO = 0.1, 0.5, 1 %	Fs = ~378, ~393, ~364	[38]
Epoxy+ Glass fiber	2Si + GO= 0.1,0.5 %	Fs = ~369, ~ 358	[39]
Epoxy+ Glass fiber	BGO = 0.3,0.6,1	Fs = ±321, ±380, ±402	Current research

*Comments 4: The conclusion part needs improvement to make it more informative to the readership.*

*Response: Conclusion section has been modified as per reviewer recommendation*

Bio-reduced graphene was synthesized by *Abutilon indicum* leaf extract, which is used as filler material to improve the mechanical and flame retardancy of epoxy matrix. The present study on bio-reduced graphene revealed the potential reduction property of *Abutilon indicum* leaf extract, which enlightened the novel pathway for its use in field of composite synthesis. Addition of BGO results in better improvement in mechanical properties viz. 28% improvement in tensile and 80 % impact behaviour of composite. It has been observed that incorporation of 0.6 wt. % BGO improves the fracture toughness of composite up to 36% and 55% of flexural strength. Flame retardancy studies revealed that V1 rating was attained for 1 wt. % BGO incorporated in matrix material and decrease in BGO wt. % reduce UL 94 rating. The developed composite better mechanical and stability to flame thus it might be suggested for potential applications such as roofing sheet and car dash boards.

## Reviewer #2

*Comment 1) Too many english mistakes need to be corrected, i suggeste to the authors to carrefully check and update the text.*

**Response:** *English of the manuscript has been polished throughout the manuscript*

*Comment 2) The authors do not provided some AFM images which can give an idea about the dispersion of the filler in the polymer matrix. It's possible to provide the images?*

**Response:** *Due to COVID pandemic situation the research labs are still remains closed. Due to this fact we are unable to carried out AFM analysis*

*Comment 3) there is a lack of references in the discussion part, the authors to compare their work with the other previous works*

**Response :** *As per reviewer advice citations has been added in discussion part and comparison tables has been as added in this updated manuscript*

**Table 1 Comparison of tensile strength with available literatures**

Matrix	Filler	Tensile strength (Mpa)	Reference
Epoxy+ Glass fiber	Nano graphene = 1, 2 %	Ts = ~78,~168	[37]
Epoxy+ Glass fiber	BGO = 0.3,0.6,1	Ts = $\pm 112, \pm 121, \pm 128$	Current research

**Table 2 Comparison of flexural strength with available literatures**

Matrix	Filler	Flexural strength (Mpa)	Reference
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Epoxy+ Glass fiber	GO = 0.1, 0.5, 1 %	Fs = ~378, ~393, ~364	[38]
Epoxy+ Glass fiber	2Si + GO= 0.1,0.5 %	Fs = ~369, ~ 358	[39]
Epoxy+ Glass fiber	BGO = 0.3,0.6,1	Fs = $\pm 321, \pm 380, \pm 402$	Current research