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Research article

# Innovative high-density polyethylene/waste glass powder composite with remarkable mechanical, thermal and recyclable properties for technical applications

W[a](#page-0-0)gih A. Sadik<sup>a</sup>, A[b](#page-0-1)del-Ghaffar M. El-Demerdash<sup>a</sup>, Abdelrahman E.A. Abokhateeb<sup>a,b,[\\*\\*](#page-0-2)</sup>, Noha A. Elessawy<sup>[c](#page-0-3),\*</sup>

<span id="page-0-0"></span><sup>a</sup> Department of Materials Science, Institute of Graduate Studies and Research, Alexandria University, Egypt

<span id="page-0-1"></span><sup>b</sup> Laboratories and Research Department, The Egyptian Ethylene and Derivatives Company (ETHYDCO), Egypt

<span id="page-0-3"></span><sup>c</sup> Advanced Technology and New Materials Research Institute, City of Scientific Researches and Technological Applications, New Borg El-Arab City, Alexandria, 21934, Egypt

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#### ABSTRACT

Several reinforcement materials are incorporated into a polymeric matrix to improve the mechanical properties and reduce the cost of the obtained composites. In this work, recycled high-density polyethylene/waste glass powder composites, compatibilized with maleic anhydride-grafted polyethylene, were prepared using a two-roll mill and compression molding techniques. Four levels of waste glass powder, 2, 10, 20 and 30% by weight, and five levels of the compatibilizer, polyethylene grafted with maleic anhydride (0.5, 1.5, 2.5, 5 and 7.5%by weight), were used. The effect of adding waste glass powder and compatibilizer concentration on the composite's mechanical properties, such as tensile strength, tensile strain, tensile modulus and thermal properties was studied. The results showed that superior mechanical properties were obtained and that the tensile strength and modulus increased with increasing waste glass powder content and compatibilizer concentration by 20 and 1.5 wt%, respectively. However, the elongation at the break decreased with the increase in both factors. The composite, which was prepared under ideal conditions, has high thermal stability and can be easily recycled and reprocessed for five cycles with high mechanical properties. This study recommends that the prepared composite, under optimum conditions, can be used as a cost-effective automobile dashboard material.

#### 1. Introduction

Thermoplastic composites are wonder materials that can be defined as hybrids of materials consisting of two phases: the polymeric matrix phase holds the dispersed phase (reinforcement phase) and shares a load with this phase; however, the two phases could be different in chemical and physical properties and separated by a distinct interface [[1](#page-10-0),[2](#page-10-1)]. The strength of the produced composite mainly depends on the ratio between the polymer matrix and the added material in the composite content [\[1](#page-10-0)]. Thermoplastic composites are increasingly important because of their advantages, such as light weight, high fatigue strength, corrosion resistance and electrical insulation [[3](#page-10-2)]. There are many different thermoplastic matrix composites used in the automotive industry, and the most common are

thermoplastic polyurethane (TPU), polypropylene (PP), polyethylene (PE) and polystyrene (PS).

High-density polyethylene (HDPE) is a thermoplastic that has a relatively low tensile modulus, low density and more uses in the automotive industry as a composite due to its lighter weights than the materials traditionally used, which positively affects fuel consumption.In addition, HDPE manufacturing is much simpler and faster than that of other types of materials. Moreover, HDPE has a long shelf life and lower environmental impact [[4](#page-10-3)] because it may come from recycled plastic. Additionally, vehicle parts manufactured with HDPE can be repaired easily, making it cost-effective.

Using composite reinforcement materials in particle form, such as carbon  $[5,6,7]$  $[5,6,7]$  $[5,6,7]$  $[5,6,7]$  $[5,6,7]$ , ceramic  $[8,9]$  $[8,9]$  $[8,9]$  or glasses particles  $[10,11]$  $[10,11]$  $[10,11]$ , is less effective in strengthening than using reinforcement materials in fibrous form, but

<span id="page-0-2"></span>\* Corresponding author. \*\* Corresponding author.

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*c* CellPress

E-mail addresses: [chem3bdoo@gmail.com](mailto:chem3bdoo@gmail.com) (A.E.A. Abokhateeb), [nony\\_essawy@yahoo.com](mailto:nony_essawy@yahoo.com) (N.A. Elessawy).

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they do improve stiffness, strength and toughness.In addition, they have the benefit of being low cost and are easier to produce and form compared to fiber-reinforced materials [\[11\]](#page-10-10). However, the advantage of using glass waste as a reinforcing material is mainly related to its low cost, high availability and thermal stability. The problem facing the manufacture of thermoplastic glass powder composites is their poor compatibility with the thermoplastic polymer, and the only way to improve the compatibility between the two phases is to select a coupling agent that reacts with both glass powder and the polymer matrix. The most broadly used coupling agent "compatibilizers" are maleated polyolefins [[12,](#page-10-11)[13,](#page-10-12)[14](#page-10-13)]. They are typically made by grafting maleic anhydride onto the polyolefin backbone via chemical interactions [[14,](#page-10-13)[15\]](#page-10-14). These linked malleated polyolefins can form many chemical bonds at the preparation temperature with hydroxyl groups on the glass particle surface [[12\]](#page-10-11), thereby strongly coupling the glass particles to the polymeric matrix [\[16](#page-10-15)].

To date, numerous approaches have been extensively developed to make polymeric composites sustainable [[17,](#page-10-16)[18\]](#page-10-17), whereas interest in waste recycling and disposable plastics has been developed in more recent decades. This work contributes to sustainable waste material processing with multiscale and multifunctional issues, such as low heating energy consumption for glass waste upcycling using a simple and reproducible approach. In addition, this research was conducted to develop a "green polyethylene composite", which can be achieved by adding certain amounts of waste glass powder (WGP), up to 20 wt %, into a recycled HDPE (RHDPE) matrix in the presence of polyethylene grafted maleic anhydride (MAgPE) as a compatibilizer using a novel procedure, and an investigation of the mechanical properties and thermal stability of RHDPE/MA-g-PE/WGP composites was carried out.

#### 2. Materials and methods

#### 2.1. Materials

Recycled high density polyethylene (RHDPE) grade material with density 0.953 g/cm $^3$ , commercial grade waste soda lime glass (WGP), and maleic anhydride-grafted-polyethylene (MAgPE) compatibilizer, also termed malleated polyethylene or polymer modifier grade (Fusabond E100), were obtained from DuPont & Industrial Polymer, Geneva, Switzerland.

### 2.2. Methods

HDPE waste bottles were cut and ground to powder using a rotary mill (Model 4 Wiley Mill, 115 V, 50/60 Hz, Wiley Standard, USA)to increase the homogeneity in particle size for both the glass waste powder and the compatibilizer. Pieces of waste glass were collected from broken house glass, washed with water and detergent, rinsed with ultrapure water and then dried at  $20 \pm 2$  °C. The composition was then determined using an X-ray fluorescence spectrometer (XRF) (ARL-XRF, Thermo Fisher, USA), as illustrated in [Table 1.](#page-1-0)

After drying, the glass pieces were ground using an Emax High Energy Ball Mill (Retsch, Germany) to achieve fine powder. The size of the fine glass powder was measured using scanning electron microscopy (SEM, JEOL, Model JSM 6360 LA, Japan), and an N5 submicron particle size analyzer (Beckman Coulter,USA) was used to evaluate the particle size distribution in addition to the particle size distribution of RHDPE and MAgPE, as shown in [Figure 1.](#page-2-0)

Thermal compounding was prepared, as shown in [Figure 2](#page-2-1), using a two roll mill machine (Prep-Mill PME 2002, Brabender CWB, USA) for 12 min at 150 °C. The sequences of material addition was such that RHDPE was added first for 3 min and then, after starting to melt, WGP was slowly added for another 3 min. Finally, the compatibilizer MAgPE (its physical properties illustrated in [Table 2\)](#page-2-2) was added to obtain homogeneity between all components and then, the produced composite paste was shaped as a sheet.

The prepared composite samples with different ratios of the components are illustrated in [Table 3.](#page-3-0) RHDPE/MAgPE/WGP composite sheet was produced with dimensions of 200 mm  $\times$  200 mm x 3 mm using a hydraulic hot press machine (Model G30H-12, Wabash Genesis, USA) at a temperature of  $160 \pm 10$  °C and pressure of 7.355 MPa for 10 min.

#### 2.3. Characterization

Surface morphology images were obtained by scanning electron microscopy (SEM, JEOL, Model JSM 6360 LA, Japan). Fourier transform infrared (FTIR) spectroscopy was utilized to investigate the chemical bonding in raw materials and composite samples using an ATR-FTIR spectrometer((Nicolet iS50, Thermo Fisher Scientific, USA). The spectra were recorded over the wavenumber range of  $400-4000 \text{ cm}^{-1}$ . The thermal stability of the prepared composites was explored by thermogravimetric analysis (TGA) using a Shimadzu TGA-50 (Japan). The analysis was conducted in a nitrogen atmosphere from room temperature to 800 °C with a heating rate of 10 °C/min. X-ray diffraction (XRD) was utilized to investigate the structure of RHDPE, MAgPE and WGP by using XRD (D2 Phaser, Bruker, Germany), while an N5 Submicron Particle Size Analyzer (Beckman Coulter, USA) was used to evaluate the particle size distributions. The mechanical properties were determined five times, according to ASTM D 638-14 for a type IV dumbbell shaped specimen ([Figure 3](#page-3-1)), for each composite sample using an Instron universal testing machine (USA) with a load cell of 5 kN at a speed of 1 mm/min for the tensile modulus and a speed of 5 mm/min for the tensile and elongation tests.

#### 3. Results and discussion

### 3.1. Mechanical properties analysis

The mechanical properties, such as tensile strength, elongation at break and tensile modulus, of the prepared RHDPE/WGP composites with varying compatibilizer contents (0.5, 1.5, 2.5, 5 and 7.5 wt. %) and four different glass reinforcement particle percentages (5, 10, 20 and 30 wt. %) were determined and illustrated in [Table 4](#page-4-0).

According to the obtained results, the tensile strength and tensile modulus of the prepared RHDPE/WGP composite without a compatibilizer, as in samples  $S_{5,0}$ ,  $S_{10,0}$ ,  $S_{20,0}$  and  $S_{30,0}$ , decreased with increasing glass reinforcement particle content, as shown in [Figure 4.](#page-4-1) This result could be attributed to the imperfect adhesion between the polymer matrix and glass particles [[19\]](#page-10-18), which led to an aggregation of glass particles (confirmed from SEM images in the next section) and that in turn made the reinforcement particles unable to support stress transferred from the polymer matrix [\[10](#page-10-9)[,20](#page-10-19)]. Furthermore, the concentration of stress at the

<span id="page-1-0"></span>Table 1. Composition of soda lime glass powder.



<span id="page-2-0"></span>

Figure 1. Particles size distribution of (a) WGP, (b) RHDPE and (c) MAgPE.

<span id="page-2-1"></span>

Figure 2. Preparation steps for RHDPE/MAgPE/WGP composite.

particle-matrix interface region leads to weakness of the particle-matrix interaction and consequently reduces the tensile strength [[21\]](#page-10-20).In other words, the tensile strength is a function of the surface contact area and the interfacial strength, the modulus is a function of the surface contact area, and filler agglomeration affects both factors. However, the significant difference of the tensile modulus of samples  $S_{0,0}$  and  $S_{5,0}$  is not very large, but visible, while no statistically significant difference between the modulus of samples  $S_{10,0}$ ,  $S_{20,0}$  and  $S_{30,0}$  is visible. The elongation at the breaks of all composite samples, with and without compatibilizer, decreased by increasing the WGP content in the polymeric matrix, which may be explained by the fact that WGP has a high rigid stiffening effect with low deformation characteristics, which acts as a stress concentrator that increases crack initiation, and consequently, the ductility of the composite will be reduced [\[22](#page-10-21)[,23](#page-10-22)].

However, to study the effect of adding MAgPE to the RHDPE/GWP composite at a constant weight percent of WGP (5 wt%), it was noticed that the tensile strength and modulus increased from 17.92 MPa and 645.18 MPa, respectively, for the uncompatibilized sample  $S_{5,0}$ , to 21.8 MPa and 963.53 MPa, respectively, for sample  $S_{5,1.5}$ , with a compatibilizer percentage of 1.5 wt. %, while further addition of compatibilizer decreased the tensile strength and modulus, as shown for samples  $S_{5,2.5}$ ,  $S_{5,5}$  and  $S_{5,7.5}$ . The tensile strength and tensile modulus,as shown in [Figure 5 \(a and c\)](#page-5-0),of all sample sets (at fixed

loading of WGP) were improved by adding MAgPE starting from 0.5 wt. % to achieve its maximum value at 1.5 wt. %, which may be because the addition of MAgPE increases the interfacial adhesion between the WGP particles and the RHDPE matrix, which reduces the interfacial stress concentration and reinforcement particle agglomeration [\[22,](#page-10-21)[24](#page-10-23)]. On the other hand, addition of MAgPE beyond 1.5 wt. % reduced the tensile strength of all sample sets, which may be explained by the formation of more hydrogen bonds between the hydrophilic sites of MAgPE and the surface hydroxyl groups on WGP particles leading to more aggregation of glass particles, subsequently decreasing the tensile strength of the composites [[25\]](#page-10-24). The best result obtained for the tensile strength was that with 20% WGP loading using

<span id="page-2-2"></span>

# <span id="page-3-0"></span>Table 3. Samples matrix composite in weight (%).



1.5%MAgPE (optimum composite sample), as shown in [Figure 5\(](#page-5-0)b), whereas the tensile strength for this optimum composite sample was 84.59% higher than that of the uncompatibilized composite sample and 29.26% higher than that of the raw RHDPE. In addition, the tensile modulus was 159.59% higher than that of the uncompatibilized composite sample and 126.78% higher than that of the raw RHDPE.

<span id="page-3-1"></span>On the other hand, the elongation at break of RHDPE/MAgPE/ WGP for all sets of composites slightly decreased with the addition of MA-g-PE up to7.5%, as shown in [Figure 5\(](#page-5-0)d), which may be due to the greater glass particle-matrix interaction, reducing the mobility of polymer chains [[22](#page-10-21)[,25\]](#page-10-24).

#### 3.2. Morphological properties analysis

The specimens submitted to mechanical tests were subjected to surface morphological investigation using SEM, as shown in [Figure 6](#page-6-0), to confirm that the best mechanical properties of the prepared sample composites were a result of good particle matrix interfacial adhesion. [Figure 6 \(a and b\)](#page-6-0) shows SEM images of the raw materials used to prepare the composites, while [Figure 6\(](#page-6-0)c, e, g and i) shows SEM images of the RHDPE/WGP composites without the MAgPE compatibilizer, in which the glass powder particles are highly aggregated and have a nonhomogeneous dispersion into the polymer matrix, which indicates poor adhesion between the polymeric phase and reinforced phase. However, by adding MAgPE, the interfacial adhesion between the glass powder and polymer matrix was



Figure 3. Sample dimension according to ASTM D 638-03.

<span id="page-4-0"></span>Table 4. The effect of adding different wt. % of WGP with different wt. % of MAgPE on the tensile strength, tensile modulus and elongation at break of RHDPE/MAgPE/ WGP composites.



<span id="page-4-1"></span>\*Sxy with x an index which represents the amount of WGP and y an index which represents the amount of compatibilizer.



Figure 4. The effect of WGP content on tensile strength, tensile modulus and elongation at break of RHDPE/WGP composites without compatibilizer.

<span id="page-5-0"></span>

Figure 5. (a) The effect of MAgPE and WGP loading on tensile strength and (b) 3D plot of this effect; (c) the tensile modulus and (d) elongation at break of RHDPE/ MAgPE/WGP composites with different loading of MAgPE and WGP.

improved, as shown in [Figure 6](#page-6-0)(d, f, h and j), which has an important role in the determination of composite properties [\[26](#page-10-25)]. The reduction in glass particle aggregation is due to an interaction between the surface hydroxyl groups of glass and the carbonyl groups of MAgPE attached to the polymeric matrix, which enhances the mechanical properties of the composites [[24](#page-10-23),[27](#page-10-26)[,28\]](#page-10-27). In comparison with the different concentrations of the reinforced waste glass powder, the size of aggregation increased with increasing glass phase content in the polymeric matrix phase, which agrees with previous studies [\[29](#page-10-28)[,30](#page-10-29)[,31\]](#page-10-30).

# 3.3. Structural properties analysis

The characteristic FTIR spectra of the raw components are presented in [Figure 7 \(a\)](#page-7-0) and Tables [5](#page-7-1), [6](#page-7-2), and [7.](#page-7-3) Polymer-compatibilizerreinforcement particle interactions were investigated and are represented in [Figure 7 \(b\),](#page-7-0) which shows the FTIR spectra of the prepared composite samples with 5, 10, 20 and 30 wt % WGP and 1.5 wt % MAgPE.

The FTIR spectrum of the prepared RHDPE/MAgPE/WGP composites showed a band of C=O stretching vibration, which belonged to MAgPE in the composite and was shifted to a lower value at  $1642 \text{ cm}^{-1}$  compared to that of pure MAgPE. This suggests that the  $C=O$  and  $C-O-C$  groups of maleic an hydride in MAgPE interacted with the hydroxyl groups on the WGP surface by hydrogen bonding [[25,](#page-10-24)[32\]](#page-10-31), which improved both the compatibility between the polymer matrix and glass particles, which in turn increased the degree of dispersion and interfacial adhesion. Furthermore, it was observed that the Si–O–Si band at 1050 – 1150cm-1 for WGP was replaced by a Si–O–C band at 800–1150  $cm^{-1}$  in the prepared composites, and this band became broader and stronger by increasing the WGP content, which confirmed that the surface group of silica changed from Si–OH to Si–O–C and that the compatibilizer was tightened chemically to the WGP surface.

[Figure 7 \(c\)](#page-7-0) shows a suggested schematic diagram of the interaction that occurred between the RHDPE/MAgPE/WGP functional groups and the formation of the composite. XRD measurements were conducted to study the structure of the RHDPE, WGP and MAgPE raw materials in addition to the RHDPE/MAgPE/WGP composite at the optimum mixture (78.5 wt.% RHDPE,1.5 wt.% MAgPE and 20 wt.% WGP) before and after the thermal milling consolidation process, as shown in [Figure 7 \(d\)](#page-7-0). As shown, the two characteristic diffraction peaks of HDPE at  $2\theta = 21.4^{\circ}$ and  $23.9^{\circ}$  corresponding to the 110 and 200 orthorhombic crystalline planes [[33\]](#page-10-32), respectively, were observed in RHDPE, suggesting that the crystal structure of RHDPE remained unchanged after cold milling. Similarly, the amorphous structure of WGP and semicrystalline structure of MAgPE were not affected by milling. However, from the XRD pattern of the blended mixture before thermal milling, a clear decrease could be seen in the intensity of the two characteristic peaks of RHDPE at the same angles due to the addition of 20 wt.% amorphous WGP. On the other hand, the XRD pattern of the blended mixture after thermal milling showed a further decrease in the intensity of the two characteristic peaks; in addition, they became broader, which may be due to the heterogeneous intercalation of WG nanoparticles between the interlayer spacing of RHDPE [\[34](#page-10-33)[,35](#page-10-34)] and chemical interaction between composite components [[25,](#page-10-24)[32\]](#page-10-31).

### 3.4. Thermal properties analysis

The thermal stability behavior of the RHDPE/MAgPE/WGP composites with the optimum compatibilizer concentration of 1.5 wt.% MAgPE and with different loading contents of WGP compared to that of raw RHDPE is shown in [Figure 8](#page-8-0)(a). The thermal degradation of all composite samples showed a single degradation step similar to raw RHDPE, and the thermal stability of RHDPE was increased by adding WGP and MAgPE, whereas MAgPE enhanced the interaction between the surface of glass

<span id="page-6-0"></span>

Figure 6. SEM images of (a) raw RHDPE, (b) raw WGP, RHDPE/WGP composites without compatibilizer (c) 5 wt% (e) 10 wt% (g) 20 wt% and (i) 30 wt% WGP content and RHDPE/MAgPE/WGP composites with 1.5 wt.% MAgPE as a compatibilizer where (d) 5 wt%, (f) 10 wt%, (h) 20 wt% and (j) 30 wt% WGP content.

particles and RHDPE to create a shield that physically protected the composite from the effects of heat. The thermal stability increases linearly with increasing glass powder content, which may be because the specific heat of glass is much less than that of RHDPE and the presence of glass powder with MAgPE enhances the thermal stability of RHDPE and decreases its volatilization [[25\]](#page-10-24)because the composite preferentially absorbs heat energy.

The temperatures obtained at mass losses of 10%, 20%, 50% and 70% are shown in [Figure 8\(](#page-8-0)b) and their values are illustrated in [Table 8.](#page-9-0) These results show a shift in the degradation temperatures with increasing WGP content from 5 wt. % to 30 wt. % and that the residue left after combustion at 800 °C for the composite that contains 30 wt. % WGP is 29.87%, which is clearly an indication for the complete degradation of MAgPE and RHDPE at this temperature and that the residue is mainly related to WGP.

<span id="page-7-0"></span>

Figure 7. FTIR spectrums of (a) raw materials (RHDPE, MAgPE and WGP), (b) RHDPE/MAgPE/WGP composites with different glass loading content and 1.5 wt. % of MAgPE, (c) suggested schematic diagram for the interaction occurred between RHDPE/MAgPE/WGP functional groups, (d) XRD patterns of RHDPE, WGP, MAgPE milled raw materials and RHDPE/MAgPE/WGP composite at optimum mixture (78.5wt.% RHDPE,1.5 wt.% MAgPE and 20 wt.% WGP) before and after thermal milling process.

<span id="page-7-1"></span>

due to polyethylene crystallinity.

# <span id="page-7-2"></span>Table 6. IR peak assignments of WGP.



# 3.5. Recyclability

Developing recyclable thermoplastic composites is required to increase the lifetime of the existing polymer materials in addition to minimizing polymer waste, which is ever increasing. To evaluate the recyclability and reprocessability, which are important factors to estimate the cost effectiveness of the prepared RHDPE/MAgPE/WGP composite with an optimum content of WGP (20 wt. %) and MAgPE(1.5 wt.%), samples that experienced mechanical fracture during testing were

# <span id="page-7-3"></span>Table 7. IR peak assignments for MAgPE.



<span id="page-8-0"></span>

Figure 8. (a) Thermal decomposition and (b) thermal stability of RHDPE/MAgPE/WGP composites with different glass loading content at fixed 1.5 wt.% of MAgPE depicted by TGA analysis.

collected, cut into small strips and then remelted using a two roll mill machine for 12 min at 150  $^{\circ}$ C. The produced composite paste was reshaped as a sheet with dimensions of 200 mm  $\times$  200 mm x 3 mm using a hydraulic hot press machine at 160 °C and pressure equal to 30 tons for 10 min. The tensile strength, elongation at break and tensile modulus were measured for dumbbell shaped specimens of each reprocessed sample. After five reprocessing cycles, it was found that the variations in the tested mechanical properties were quite small after each cycle, as shown in [Figure 9](#page-9-1). For instance, the differences between the first cycle and the last cycle were quite small (1.05 MPa, 1.5 MPa and 0.98% for tensile strength,



<span id="page-9-0"></span>Table 8. The degradation temperatures  $T_{10\%}$ ,  $T_{20\%}$ ,  $T_{50\%}$  and  $T_{70\%}$  of RHDPE/MAgPE/WGP composites with different glass loading content and 1.5 wt.% of MAgPE depicted by TGA analysis.

<span id="page-9-1"></span>



tensile modulus and elongation at break, respectively), which is a good indication of RHDPE/MAgPE/WGP composite structure stability. Furthermore, there was an extremely small increase in the values of the measured mechanical properties for the second recycling cycle. However, the oddity in the second cycle may be due to the components of the composite having a homogeneous distribution due to remixing, which slightly enhanced the mechanical properties, but due to stress accumulation, the mechanical properties then decreased in the following cycles.

# 4. Conclusion

Novel, low-cost, and fully recyclable thermoplastic composites were produced using a simple and environmentally conscientious approach by reinforcing recycled high-density polyethylene waste with waste glass powder in the presence of maleic anhydride-grafted polyethylene as a compatibilizer. The mechanical evaluation showed that the tensile strength, tensile modulus and elongation at break of the prepared composites were affected by the reinforcement glass powder and compatibilizer content, whereas the higher the load was, the higher the modulus up to the optimum condition at 20 wt. % waste glass powder and 1.5 wt. % MAgPE, while the evaluation of the thermal properties showed high thermal stability for the optimum composite. Recycling of the prepared composite did not affect its mechanical and thermal properties. Therefore, this method holds great promise to replace high-cost polymer-based composite automotive components to achieve more cost-competitive materials for the future.

#### **Declarations**

#### Author contribution statement

Wagih A. Sadik, Abdel-Ghaffar M. El-Demerdash: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

Abdelrahman E. A. Abokhateeb: Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Noha A. Elessawy: Analyzed and interpreted the data; Wrote the paper.

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#### Data availability statement

Data included in article/supplementary material/referenced in article.

#### Declaration of interests statement

The authors declare no conflict of interest.

#### Additional information

No additional information is available for this paper.

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