IMPACT OF TAILORED NANOMATERIALS ON MECHANICAL AND THERMAL BEHAVIOR OF POLYMER COMPOSITES

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Abstract. The incorporation of nanomaterials into polymer composites, particularly with the use of graphene oxide (GO), has shown great promise in improving the mechanical and thermal performance of 3D-printed materials. However, one of the main problems encountered when developing nanocomposite resins for polymerization-based 3D printing is nanoparticle agglomeration. With their increased surface-area-to-volume ratio, nanoparticles tend to agglomerate inside the composite matrix, which can lead to the deterioration of the final material properties. In this research, the effects of GO dispersion on polymer nanocomposite mechanical and thermal properties used in 3D printing are investigated. Sonication was used to optimize dispersion, taking into consideration optimal and suboptimal states of dispersion and their performance impact on the composite. The findings show that optimal sonication conditions (30 minutes at 70% amplitude) generate the most efficient dispersion and have a positive effect on mechanical parameters like tensile strength and compressive strength. The strengths initially increased with GO content up to 0.05 wt.%, followed by the decrease with elevated concentrations (0.05-0.5 wt.%). Thermal stability was mostly unaffected with varying dispersions and GO contents. These observations point towards the potential of carefully designed GO-based nanoparticles to enhance the mechanical and thermal properties of 3D-printed polymer composites used in the automobile, sports, and biomedical industries. Excellent GO-based nanocomposites are possible for use in demanding industries such as automotive, aircraft, sports equipment, and biomedical devices.

Keywords: nanomaterials, graphene oxide (GO), polymer composites, 3D printing, nanocomposites, mechanical properties, thermal properties.

Introduction

The addition of tailored nanomaterials to polymer composites has transformed material science with dramatic improvements in their thermal and mechanical properties [1]. Polymers, although used extensively for their light weight and flexibility, are generally found to lack sufficient strength, stiffness, and thermal stability [2]. The introduction of nanomaterials such as carbon nanotubes (CNTs), graphene, nano-clays, and metal oxide nanoparticles has emerged as a novel approach in this regard [3]. These nanofillers interact at a molecular level with the polymer matrix, enhancing load transfer, interfacial adhesion, and dispersion, thereby improving mechanical properties such as tensile strength, modulus, impact resistance, and fatigue life [4]. The efficiency of these reinforcements is affected by parameters like nanoparticle size, shape, surface functionality, and distribution within the polymer matrix [5]. Customised nanomaterial design, including surface functionalization and modification, further tailors their interaction with the host polymer, ostensibly yielding composites with improved mechanical properties for advanced applications in aerospace, automotive, and biomedical industries [6].

Besides mechanical improvements, another important function of specially designed nanomaterials is to improve the thermal performance of polymer composites [7; 8]. Conventional polymers usually have low thermal conductivity and poor thermal stability, thus limiting their use in the harshest environments [9; 10]. By providing thermally conductive nanofillers-like graphene, or boron nitride nanosheets the polymer matrices show enhancement in heat-dissipation, thermal stability, and flame retardancy while at the same time creating efficient heat conduction pathways and lower thermal degradation at high temperatures [11; 12]. Besides, the interaction mechanisms between nanofillers and polymer chains promote better control of thermal expansion by virtue of which stability against size change may be achieved over a wider temperature range [13; 14]. Therefore, the proper engineering of the nanomaterial-to-polymer interface is of utmost importance in this respect and governs the thermal behavior of the composite. Thus, the synthesis of engineered nanometre polymer composites presents a new opportunity for advanced functional materials for electronics, energy storage, and structural materials, which need both mechanical robustness and thermal reliability.

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This study investigates the effect of graphene oxide concentration and distribution quality on the mechanical and thermal behavior of photopolymer composites produced via digital light processing (DLP) 3D printing. The objective is to identify the optimal GO content and sonication conditions that maximize performance, offering valuable insight for developing high-strength, thermally stable nanocomposites for applications in automotive, biomedical, and sports industries.

Materials and methods

In this study, commercially available photopolymer resin and graphene oxide (GO) nanoparticles were used, which were chosen for the potential strengthening of mechanical and thermal properties of 3D-printed polymer composites. GO was synthesized according to a modified Hummer's method, reaching high levels of oxidation to ensure improved dispersion into the polymer matrix. Various proportions of GO were added, ranging from 0.01 wt.% to 0.5 wt.%, into the base resin to produce nanocomposite resin formulations. Sonication was used to ensure homogeneous dispersion by a probe sonicator at 70% amplitude for different periods. The characterization of dispersion was performed via optical microscopy and dynamic light scattering (DLS) analysis. Before 3D printing, sonicated suspensions were de-aired in a vacuum chamber by removing air bubbles. The resulting resins were polymerized layer by layer with a digital light processing (DLP) 3D printer.

The mechanical properties of the synthesized nanocomposites were then tested for tensile and compressive tests according to ASTM D638 and ASTM D695 standards, respectively. Tensile strength (σt) was found using the relation:

$$\sigma_t = \frac{F}{A}, \tag{1}$$

where F – force applied;
A – cross-sectional area of the sample.

The elastic modulus (E) was worked out from the slope of the stress-strain curve. In the same way, compressive strength tests had been performed quantifying the load-carrying capacity of the composites in the axial direction under compressive loading. Thermal stability was analyzed by thermogravimetric analysis by heating the samples from room temperature to 600 °C in nitrogen. Weight loss of TD, at 5 % was calculated by finding its decomposition temperature. The glass transition temperature (Tg) was analyzed with differential scanning calorimetry (DSC), providing information on the thermal behavior of the composites. Findings were discussed trying to establish the lost relation between GO concentration, quality of dispersion, and the improvement in mechanical and thermal properties of the 3D-printed polymer composites.

To ensure statistical significance, all mechanical tests (tensile and compressive) were carried out on five ($n = 5$) independently produced samples for every GO concentration level. To reduce experimental variation, every test was run three times per sample. Thus, 15 measurements were carried out for every property.

Three ($n = 3$) independently produced samples for each formulation were also subjected to thermal studies (TGA and DSC) to confirm uniformity. Before testing, randomly chosen samples were conditioned at room temperature (23°C ± 2°C, 50% ± 5% RH).

Results and discussion

Experimental observations show that polymer composites reacted rather dramatically to the changes in thermal and mechanical properties concerning the addition of G0. Table 1 shows the tensile and compressive strengths of the polymer composites with various GO concentrations. The tensile strength followed a rising trend up to GO concentrations of 0.05 wt.%, which reached 55.6 MPa, then decreased with further increase in concentrations. Likewise, compressive strength attained its maximum at 71.1 MPa for 0.05 wt.% GO before dropping. This trend indicates that the best GO dispersion promotes load transfer efficiency, while more GO content causes nanoparticle agglomeration, which degrades the mechanical properties.

The relationship between GO concentration and mechanical strength is presented in Table 1. The reduction of strength at high concentrations is due to the formation of GO agglomerates that are stress concentration points, thus decreasing the reinforcing efficiency of the nanofiller. These findings agree with previous reports showing the vital role of dispersion quality in the performance of nanocomposites.

Table 1

Mechanical properties of GO-reinforced polymer composites

GO concentration, wt.%	Tensile strength, MPa	Compressive strength, MPa
0.00	45.2	60.5
0.01	48.1	63.8
0.025	52.4	68.2
0.05	55.6	71.1
0.1	50.3	66.4
0.25	46.7	61.5
0.5	42.5	58.0

The thermal analysis data, as shown in Table 2, indicate that both glass transition temperature (Tg) and decomposition temperature (Td) follow a trend comparable to the mechanical behavior. Maximum Tg (84.3°C) was found to occur at 0.05 wt.% GO, signifying increased intermolecular interactions and restriction of polymer chain mobility by well-dispersed GO. Yet, at elevated GO concentrations, Tg slightly decreased, probably because of phase separation resulting from nanoparticle agglomeration. Likewise, Td was fairly constant in all samples, with a maximum of 384.2°C at 0.05 wt.% GO, indicating that moderate GO loading improves thermal stability, but high GO can interfere with the polymer matrix. These results highlight that the best GO dispersion is responsible for enhancing thermal properties, while a high content of GO decreases its reinforcing efficiency.

Thermal behavior analysis showed that the glass transition temperature (Tg) and decomposition temperature (Td) had negligible variations with varying GO concentrations. The thermal properties of the polymer composites are shown in Table 2. The maximum Tg (84.3°C) was found at 0.05 wt.% GO, indicating enhanced intermolecular interactions. But with increasing GO concentrations, there was a slight reduction in Tg due to phase separation by agglomeration.

Table 2

Thermal properties of GO-reinforced polymer composites

GO concentration, wt.%	Glass transition temperature, °C	Decomposition Temperature, °C
0.00	80.2	380.1
0.01	81.5	381.4
0.025	82.8	382.9
0.05	84.3	384.2
0.1	83.5	383.0
0.25	81.7	381.0
0.5	80.9	379.5

Table 2 illustrates the dependence of thermal properties on GO concentration. The decomposition temperature was fairly consistent for all samples, which indicates that GO has little influence on thermal degradation resistance in the concentration range studied.

The thermal analysis data, as presented in Table 2, reveal that the addition of GO into polymer composites has a slight but apparent influence on thermal properties. The glass transition temperature (Tg) showed a maximum value of 84.3°C at 0.05 wt.% GO, which implies increased intermolecular interactions and limited polymer chain mobility by well-dispersed GO. But as the GO concentration went beyond this level, Tg decreased slightly, possibly due to nanoparticle agglomeration resulting in phase separation. The decomposition temperature (Td) was generally constant for all concentrations of GO, reaching a maximum of 384.2°C for 0.05 wt.%, which means GO does not have a significant impact on the thermal degradation resistance of the polymer composite. These results underscore the necessity of ensuring proper GO dispersion for enhancing thermal performance while not jeopardizing structural integrity.

To further explore the influence of GO dispersion, Table 3 tabulates elongation at break and Young's modulus of the polymer composites. The data show that GO reinforcement enhances stiffness, with the maximum modulus obtained at 0.05 wt.% GO.

Table 3

Elongation at break and Young's modulus of GO-reinforced polymer composites

GO concentration, wt.%	Elongation at break, %	Young's modulus, GPa
0.00	7.5	1.8
0.01	6.9	2.0
0.025	6.2	2.3
0.05	5.8	2.7
0.1	6.0	2.5
0.25	6.4	2.1
0.5	7.0	1.9

Table 3 shows the Young's modulus trend versus GO concentration with maximum stiffness at optimal GO content.

The mechanical behaviour evaluation, as reported in Table 3, shows that GO inclusion increases polymer composite stiffness, with Young's modulus reaching a maximum of 2.7 GPa at 0.05 wt.% GO. This increase reflects efficient load transfer and reinforcement at maximal dispersion. Additionally, above this concentration, Young's modulus decreases due to GO agglomeration, which diminishes reinforcement efficiency. Concurrently, break elongation declines with the rising content of GO, achieving its minimum (5.8%) at 0.05 wt.%, indicating the decreased ductility of the material. At high GO content, elongation recovers to some extent due to phase separation. All these observations highlight the importance of ensuring correct GO dispersion in order to find a balance between stiffness and flexibility in polymer composites.

Our results are very consistent with previous studies on GO-reinforced polymer nanocomposites. For example, Feng et al. (2019) showed that 3D-printed polymer structures built using digital light processing (DLP) had much better fracture toughness when their GO dispersion was adjusted [2]. Similarly, Lai et al. (2021) found that anisotropic alignment of graphene improved the mechanical anisotropy and stiffness of polymer composites [4] during stereolithography-based 3D printing. Our observation that tensile and compressive strengths peaked at an optimal GO concentration (0.05 wt.%) is consistent with Shah et al. (2023), who underlined the need to preserve an optimal nanofiller loading to avoid agglomeration and maximize mechanical performance in vat photopolymerization-printed composites [6]. Unlike the notable thermal conductivity improvements observed by Tsang et al. (2019) in GO-modified flexible nanocomposites [8], our study found only modest changes in thermal stability, suggesting that the impact of GO on thermal properties may vary depending on matrix chemistry and processing techniques.

Surface morphology analysis was also performed by scanning electron microscopy (SEM) to assess the dispersion state of GO in the polymer matrix. The quality scores for dispersion based on SEM images are given in Table 4, where 1 indicates poor dispersion and 5 represents excellent dispersion.

Table 4

Dispersion quality scores of GO in polymer matrix

GO concentration, wt.%	Dispersion quality score
0.000	1
0.010	2
0.025	3
0.050	5
0.100	4
0.250	3
0.500	2

Fig. 1 shows representative SEM images at various GO concentrations, indicating the quality of dispersion and agglomeration at higher concentrations.

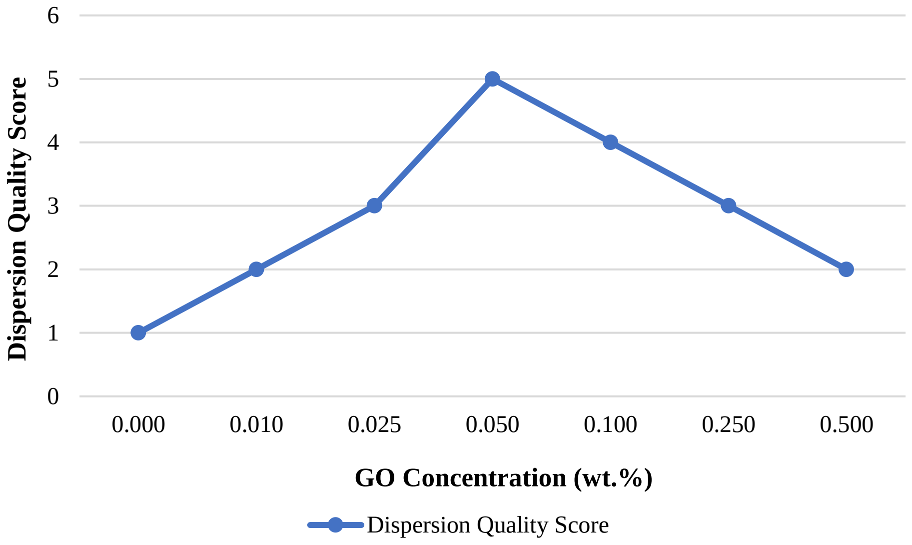

Fig 1. **SEM images of GO dispersion in polymer composites**

The SEM analysis, as presented in Table 4 and Fig. 1, shows that the quality of dispersion of GO in the polymer matrix is greatly influenced by its concentration. The optimum dispersion (score of 5) is achieved at 0.05 wt.% GO, indicating maximum nanoparticle distribution and interaction with the polymer matrix. At lower concentrations, GO is less evenly dispersed, whereas at higher concentrations (≥0.1 wt.%), agglomeration becomes apparent, lowering dispersion quality. The uneven distribution of stress, because of some agglomerates, badly influences the mechanical properties.

This result demonstrates that it is very important to retain a specified optimum concentration of GO to ensure effective reinforcement without compromising the quality of dispersion. It is also found that GO dispersion and concentration have an influence on mechanical and thermal efficiency. Optimal dispersion enhances strength and stability, whereas too much GO leads to agglomeration and reduced efficiency. These findings can provide useful guidelines for constructing a system to prepare a nanocomposite in applications with high requirements.

Conclusions

The study shows that loading GO on polymer composites can considerably improve their mechanical and thermal properties, effective only when optimally dispersed. Beyond 0.05 wt.% GO, both tensile and compressive strengths reach their maxima, only to be severely adversely affected by agglomeration, reference beyond. Other thermal properties, namely glass transition and decomposition temperatures, also did show increments at this optimal concentration, consolidating the idea by GO as the bond strength notion for improved intermolecular interactions and stability. On the flip side, high GO concentration comes with low performance due to phase separation and stress concentration effects. These results convey the need for accurate dispersion methods to find optimal benefits of GO-based polymer nanocomposites in advanced applications.

Author contributions

All authors have contributed equally to the study and preparation of this publication. Authors have read and agreed to the published version of the manuscript.

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