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Optimized Functionalization of Graphene Oxide for Enhanced Mechanical Properties in Epoxy Resin Composites

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Abstract: Epoxy resins have widespread applications across various industries, such as anticorrosive coatings, owing to their exceptional attributes. However, there is a constant demand for enhancements to their mechanical characteristics to cater to the requirements of unique and specialized conditions. In this work, graphene oxide modified by 4,4'-Oxydianiline (MGO) was prepared using a covalent grafting reaction. The MGO in epoxy resin composites exhibited a rougher morphology and thin layers with a superior tensile strength (38 MPa), elastic modulus (358 MPa), flexural strength (85 MPa), flexural modulus (957 MPa), and hardness (62 HD). The results indicated that the mechanical properties of epoxy resin are significantly improved by MGO and the improved mechanical properties of epoxy resin composites are due to the strong interfacial bonding between MGO and epoxy.

Keywords: graphene oxide; 4,4'-Oxydianiline; epoxy; mechanical properties

1. Introduction

Epoxy resin has been extensively used in aerospace [1], electronic instruments [2], engineering construction [3], and other fields [2,4] due to its superior excellent physical properties, chemical resistance, stability, and unique processability [5]. However, bisphenol A epoxy resins, the most commonly used epoxy resins, are limited by defects in their mechanical properties, such as high brittleness and poor tensile strength and flexural strength [6]. Thus, a large number of methods have been reported to improve the mechanical properties of epoxy resins, including changing the molecular structure of the matrix and adding compatible functional inorganic fillers [7,8]. Simultaneously, researchers have explored altering the structure of the curing agent, a pursuit that has garnered considerable interest. For instance, Alexey Orlov et al. [9] undertook the synthesis of curing agents featuring varying phosphazene contents. Their investigations revealed that the complex comprising 30% modifier significantly enhanced both the mechanical robustness and fire-retardant properties of the epoxy resin. However, the change of curing agent will make the operation complicated and reduce the efficiency. Among these methods, combination with the second phase of superior nanofillers has been developed as a simple and efficient method [10,11].

Due to its incredible mechanical, electrical, and thermal properties, graphene, a unique two-dimensional layered material, has generated great interest [12,13]. Cahit Orek et al. [14] solved the problem of gold(I) N-heterocyclic monocarbene-adsorbing graphene as a drug nanocarrier via calculations. Studies have shown that graphene and its derivatives can effectively improve the mechanical properties of polymers, Anastasia Konstantinova et al. [15] ascertained that the incorporation of a phosphazene-containing curing agent, coupled with thermally expanded graphite, markedly enhanced the fire resistance, as well as the physical and mechanical characteristics, of epoxy resin. However, it could only happen if graphene nanoplatelets were well dispersed in the polymer matrix [16–19]. In order to improve the dispersion of GO, Zhang et al. [20] employed atom transfers radical polymerization to link



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GO and boron nitride, subsequently incorporating this composite as a filler into the epoxy resin matrix. Their investigation revealed commendable attributes, including noteworthy thermal conductivity, electrical insulation, and compression capabilities. In this regard, surface covalent functionalization has been conducted to enhance uniform dispersion and enhance compatibility with polymer matrices [21–23]. But there are still some deficiencies because of complex modification reactions and severe conditions, which will reduce efficiency and increase costs, even causing the degradation of the polymer matrix.

Due to its excellent physical properties and corrosion resistance, epoxy resin is widely used as a film-forming material in coatings. We have previously investigated the impact of covalent and non-covalent modifications to GO on the corrosion resistance and weathering resistance of epoxy resin coatings [24–26]. In this work, we report a facile strategy to synthesize 4,4'-Oxydianiline-modified graphene oxide (MGO) via surface covalent functionalization and modified graphene oxide/epoxy resin composites (MGO/EP) via two-step synthesis. The morphology and mechanical properties of MGO/EP were compared, and the effect of MGO on the mechanical properties was investigated.

2. Experiments

MGO was synthesized using a covalent grafting reaction. Firstly, 0.25 g 4,4'-Oxydianiline and 0.5 g GO were added into 50 mL and 100 mL N,N-Dimethylformamide (DMF), respectively, and stirred for 1 h. Subsequently, the mixture was stirred and refluxed for 4 h at 80 °C and then dried in a vacuum oven at 60 °C overnight. EP/MGO were synthesized using a two-step synthesis. Firstly, MGO and EP were added into ethanol under stirring for 1.5 h in turn and then dried at 80 °C overnight. Polyamide curing agents (650) were added into EP under stirring for 0.5 h, and the mixed solution was poured into a preheated Teflon mold and cured in a vacuum oven at 80 °C overnight. Figure 1 shows the synthesis mechanism and preparation method of MGO and EP/MGO.

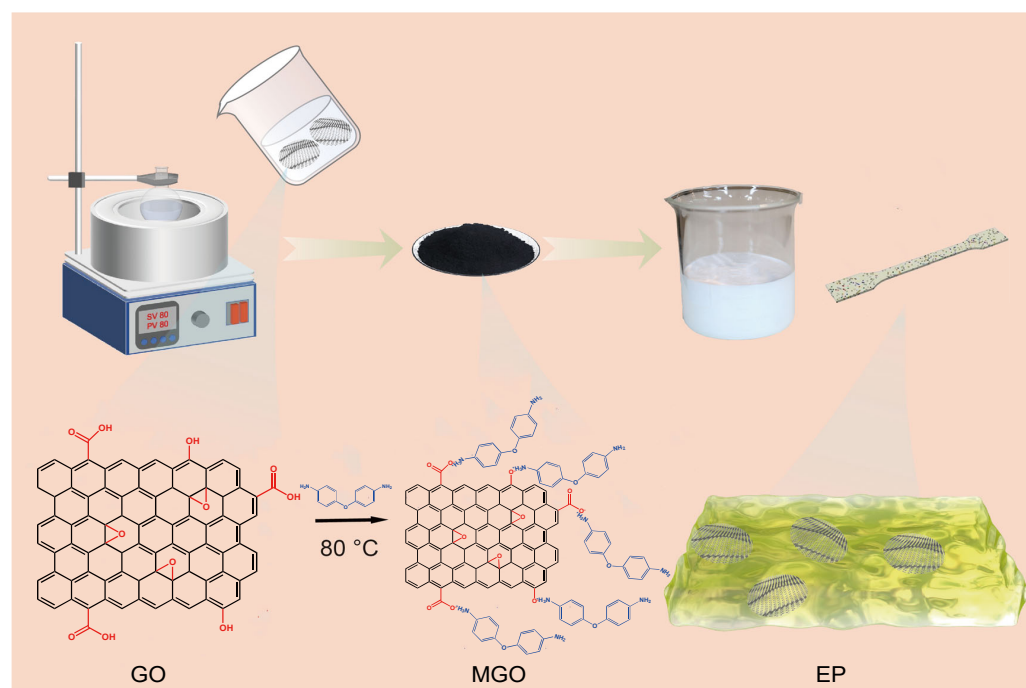


Figure 1. Schematic diagram of the preparation and the reaction between 4,4'-oxydianiline and GO.

The morphology of MGO was observed using scanning electron microscopy (SEM, FEI QUANTA 650) (FEI, Hillsboro, OR, USA) and transmission electron microscopy (TEM, JEOL 2100) (JEOL, Tokyo, Japan). The powder specimens were dispersed in ethanol and desiccated for SEM and TEM analysis. The modification of GO was evaluated using X-ray diffractometry (XRD), Fourier transform infrared (FTIR) spectrum, Raman spectrum, and

X-ray photoelectron spectrum (XPS) measurements. The GO or MGO powder specimens were promptly situated within the XRD sample chamber, and the XRD data were acquired within the angular span from 5° to 60° . FTIR was obtained using a Bruker TENSOR II spectrometer (Bruker, Karlsruhe, Germany), and the scanning range was $4000\text{--}400\text{ cm}^{-1}$. The solid powder sample was mixed with a small amount of potassium bromide powder, and the mixed powder was fined and pressed into tablets for infrared spectroscopy. Raman spectra were obtained using a Horiba IHR320 spectrometer (Horiba, Kyoto, Japan) with a scanning range of $500\text{--}4000\text{ cm}^{-1}$ and an excitation wavelength of 532 nm. The solid powder was dispersed in ethanol and dried on a glass slide for Raman spectroscopy. The powder specimens were affixed directly onto the XPS sample platform for XPS examination. The tensile and bending properties of composite materials were tested using a microcomputer-controlled electronic universal testing machine, according to GB/T 13525-1992 [27] and GB/T 6569-2006 [28] standards.

3. Results and Discussion

Upon comparing the XRD (Figure S1), FTIR (Figure S2), and Raman (Figure S3) test results, the optimal modification reaction ratio between 4,4'-Oxydianiline and GO (1:2) was determined. MGO was synthesized using this optimal ratio for subsequent research endeavors. The XRD spectra of GO and MGO are shown in Figure 2a. The characteristic diffraction peaks at 12.445° of GO and 9.074° of MGO represent the 0 0 1 reflection peak, corresponding to an interlayer spacing of about 0.709 nm and 0.973 nm, respectively. In the FT-IR spectrum of GO, the peaks at 3397 cm^{-1} , 1732 cm^{-1} , 1612 cm^{-1} , 1236 cm^{-1} , and 1059 cm^{-1} are attributed to --OH , C=O , C=C , C--O--C , and C--O bands, respectively. This shift implies a reduction in the interlayer separation relative to that observed in GO. The computed interlayer spacing value provides compelling evidence indicating the covalent grafting of 4,4'-Oxydianiline molecules onto the surface of GO, thereby corroborating the presence of such molecules [26]. Meanwhile, in the FT-IR spectrum of MGO, the C=O band nearly disappears and new bands at 1496 cm^{-1} and 823 cm^{-1} arise, representing the C--N and N--H stretch vibrations of the methylene group (Figure 2b). The Raman spectra of GO and MGO displays two peaks at 1354 cm^{-1} (D band) and 1583 cm^{-1} (G band), and the I_D/I_G value increased from 1.89 of GO to 2.84 of MGO, indicating that covalent bonding occurs between 4,4'-Oxydianiline and GO without the significant destruction of the carbon lattice [29]. Furthermore, it is proven that, subsequent to covalent bonding, the degree of disorder within MGO tends to escalate when juxtaposed with GO. Figure 2d shows the XPS spectra of GO and MGO. Compared with GO, there is a characteristic peak at N 1s originating from 4,4'-Oxydianiline, and the appearance of C--N further confirms the covalent functionalization of GO. The high-resolution C 1s XPS spectrum of GO can be meticulously deconvoluted to discern distinct bands—namely, C--C (at 284.56 eV), C--O (at 286.87 eV), and C=O (at 289.01 eV)—as shown in Figure 2e. Meanwhile, the C--C peak of MGO undergoes attenuation, and a fresh C--N peak (at 285.69 eV) emerges, as shown in Figure 2f. The observed distinctions signal the alterations to the chemical bonding and functional groups stemming from the covalent grafting modification with 4,4'-Oxydianiline.

The morphology of GO and MGO was observed and is shown in Figure 3. The diagram clearly illustrates the pronounced layered structure inherent in both GO and MGO. As shown in Figure 3a,b, there are a few folds on the surface and edges of GO, which are stacked in layers. After the modification treatment, the surface of MGO becomes looser and rougher (Figure 3c,d). Thin layers of GO and MGO can be observed in Figure 3e,f, and the corresponding selected area electron diffraction (SAED) pattern (inset in Figure 3e,f) also suggests that MGO has a more obvious crystal structure than GO.

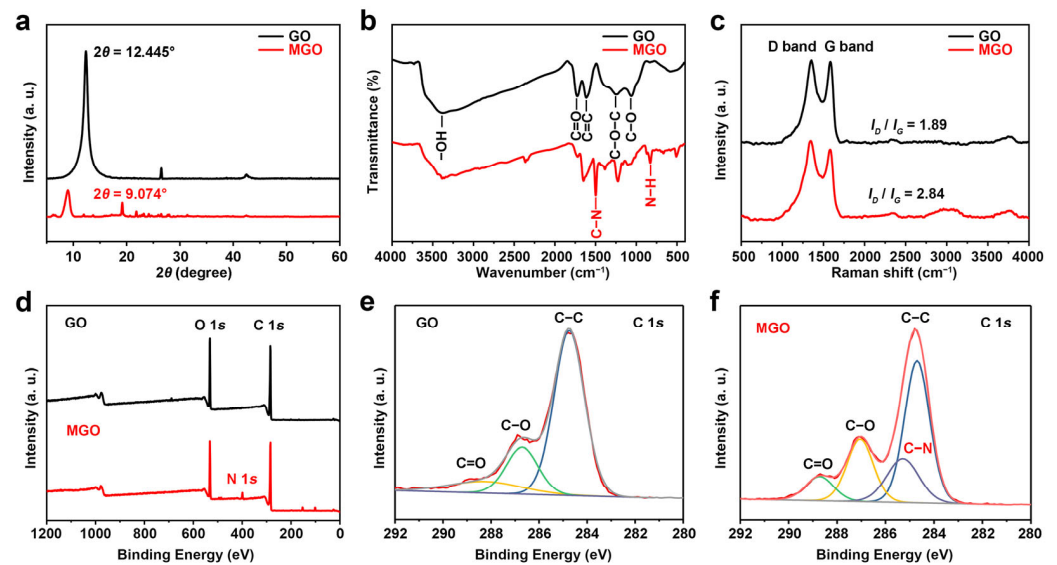


Figure 2. XRD spectra (a), FT-IR spectra (b), Raman spectra (c), and XPS spectra (d) of GO and MGO, and C 1s XPS spectra of (e) GO and (f) MGO.

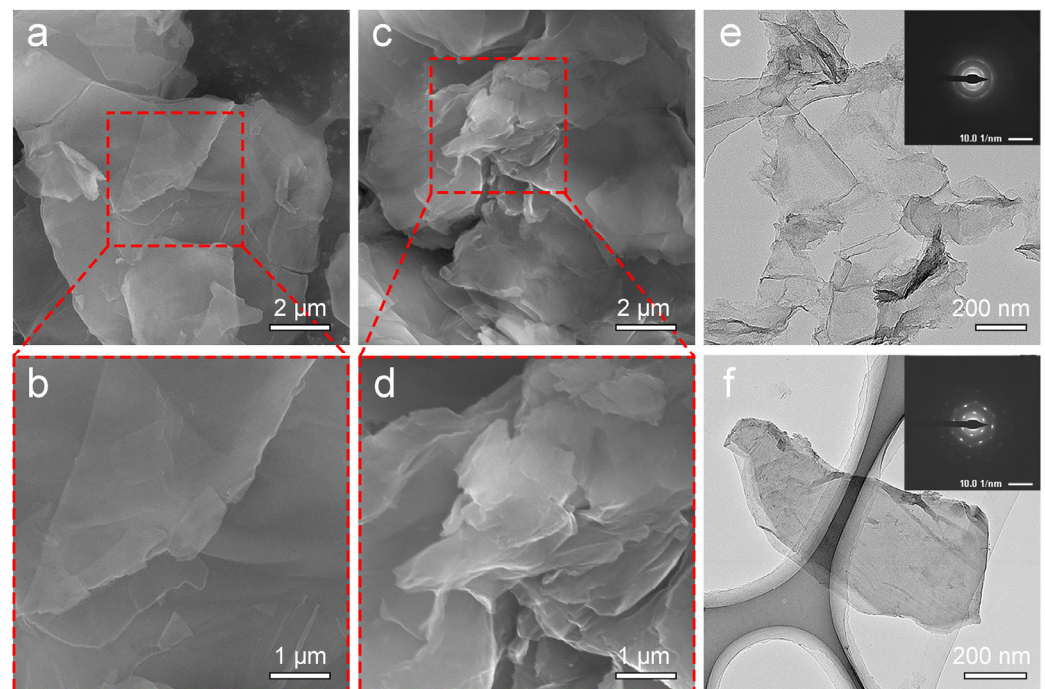


Figure 3. SEM images of GO (a,b) and MGO (c,d), and TEM images of GO (e) and MGO (f). The inset of (e,f) shows the corresponding SAED pattern.

In order to investigate the effect of MGO addition on the mechanical properties of EP/MGO, 0.1 wt.%, 0.3 wt.%, 0.5 wt.%, and 1.0 wt.% of MGO were added into epoxy resin, which was named EP/MGO01, EP/MGO03, EP/MGO05, and EP/MGO10, respectively. The tensile properties of EP and EP/MGO were tested and compared, as shown in Figure 4. The tensile strength of EP/MGO01 increased from 24 MPa to 38 MPa, and the elastic modulus increased from 248 MPa to 358 MPa. As the amount of MGO added increased, the tensile property and flexural property decreased in varying degrees, as shown in Figure 4b. Similarly, the elongation at break originally presented a significant increase and then decreased to varying degrees (Figure 4c).

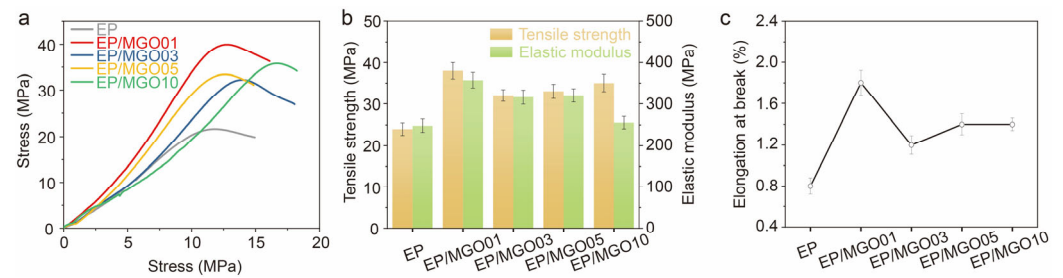


Figure 4. Tensile properties of epoxy and its composites with different MGO loadings: stress–strain curves (a), tensile strength and elastic modulus (b), and elongation at break (c).

The fractured surfaces of EP, EP/GO (the addition amount of GO is 0.1 wt.%), and EP/MGO01 were observed using SEM, as shown in Figure 5. There are small faceted features on the fracture surface of EP, indicating a brittle fracture with poor tensile strength. Likewise, the fracture surface of EP/GO retains small-faceted attributes akin to EP, suggestive of its brittle fracture nature. However, the fracture surface of EP/GO exhibits a heightened roughness compared to EP, indicative of GO incremental enhancement of EP mechanical resilience to a certain degree. Meanwhile, EP/MGO01 exhibits a relatively rough fracture surface with river-like patterns, which is a typical feature of ductile fracture. Moreover, no obvious pull-out trace can be observed, indicating the strong MGO-EP interfacial bonding as a result of the functional group of MGO, which has a significant influence on the mechanical properties of epoxy resin composites.

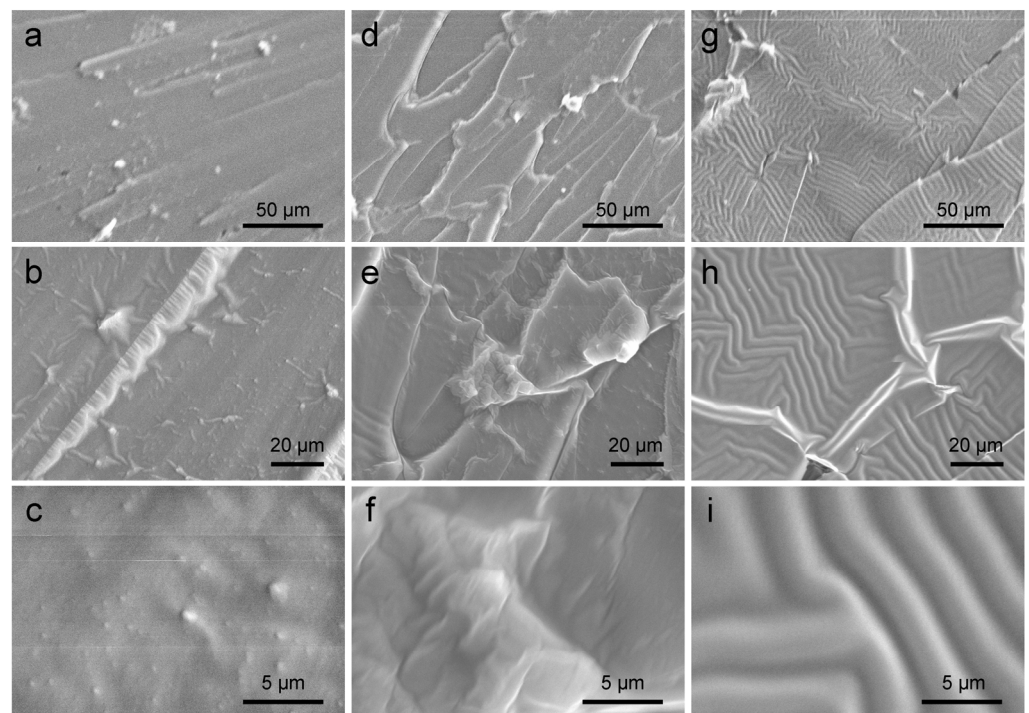


Figure 5. SEM images of the fractured surface of EP (a–c), EP/GO (GO content: 0.1 wt.%) (d–f), and EP/MGO01 (g–i).

The flexural properties and hardness of EP and EP/MGO were tested and compared, as shown in Figure 6. Figure 6a shows the typical load–displacement curves of EP and EP/MGO, and the smooth curves indicate that the cracks propagated in a controlled manner [30]. The flexural strength of EP/MGO01 increased from 48 MPa to 85 MPa, and the flexural modulus increased from 473 MPa to 957 MPa (Figure 6b). The impact strength of EP is 4.507 kJ/m², while the impact strength of EP/MGO01 is 8.462 kJ/m².

In comparison to EP, the impact strength of EP/MGO01 increased by 88% (Figure 6c). Moreover, the hardness of EP is significantly improved by MGO, as shown in Figure 6d, and the highest hardness of EP/MGO01 is 62 HD, which is 1.44 times that in EP (43 HD).

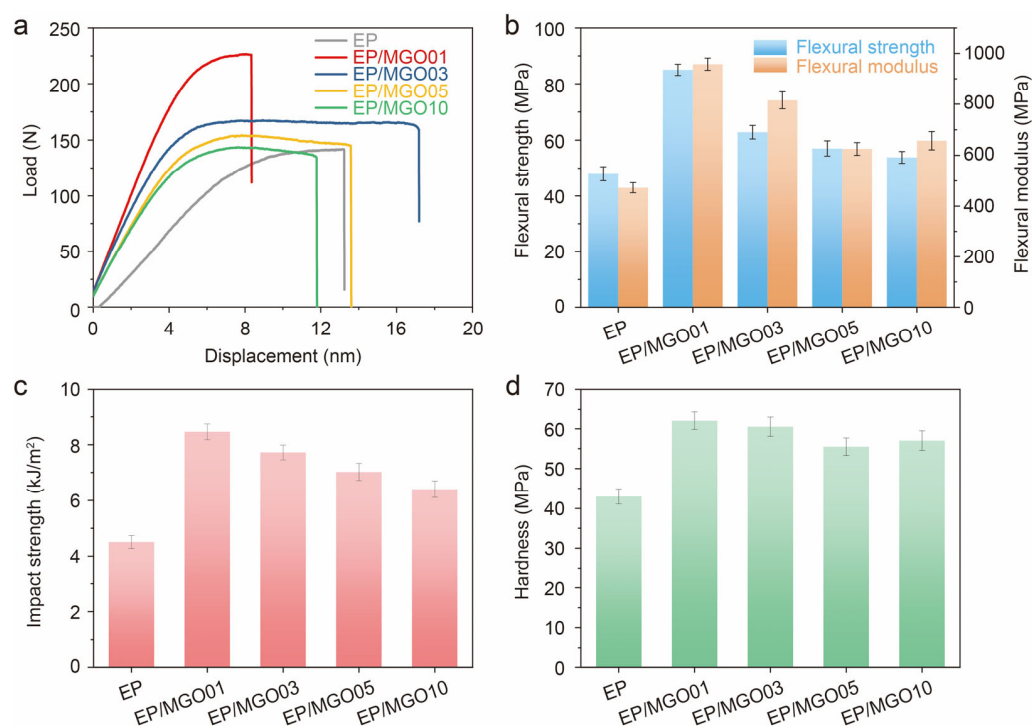


Figure 6. Load–displacement curves (a), flexural strength and flexural modulus (b), and impact strength (c) and hardness (d) of epoxy and its composites with different MGO loadings.

When MGO is evenly distributed within epoxy resin, the strong interfacial bonding between MGO and epoxy in composites offers several benefits for enhancing mechanical properties. The strong bond ensures efficient load transfer between the MGO filler and the epoxy matrix, leading to increased strength and stiffness in the composite material. And a well-bonded interface can contribute to lower wear and friction between the MGO and the epoxy matrix, resulting in improved tribological properties in the composite. Moreover, strong interfacial bonding minimizes the risk of interface delamination or debonding, thereby improving the long-term durability and reliability of the composite material. Therefore, the addition of MGO improves the mechanical properties of epoxy resin.

However, when there is an excessive amount of MGO added, the particles may agglomerate and clump together, leading to localized stress concentrations and interface breakdown, which ultimately reduce the overall performance of the epoxy matrix. And excessive amounts of MGO may weaken interface adhesion, as the reduced distance between filler particles hinders effective interactions between the epoxy resin and the filler, thereby diminishing the strength and toughness. Therefore, excessive amounts of MGO may lead to a decrease in the mechanical properties of epoxy resin, requiring appropriate control of the additive amount to achieve optimal performance. In this work, the optimal MGO addition amount is 0.1 wt.%.

4. Conclusions

MGO has been successfully fabricated via a covalent grafting reaction of GO and 4,4'-Oxydianiline. The looser and rougher surface and thin layers of MGO were confirmed using SEM and TEM. As feature-enhanced fillers, MGO in epoxy resins showed a significant increase in mechanical properties; the tensile strength of EP/MGO01 increased from 24 MPa to 38 MPa, the elastic modulus increased from 248 MPa to 358 MPa, the flexural strength increased from 48 MPa to 85 MPa, the flexural modulus increased from

473 MPa to 957 MPa, and the hardness increased from 43 HD to 62 HD. The aforementioned outcomes stem from the incorporation of MGO, which serves to ameliorate imperfections such as pores and microfractures within the EP matrix while concurrently augmenting its density. Furthermore, the introduction of MGO fortifies the cohesion at the MGO-EP interface. The results demonstrate that the MGO is very promising as an alternative effect filler candidates for high-mechanical-property composites.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/coatings14050609/s1>, Figure S1: XRD spectra of GO and MGO samples; Figure S2: FT-IR spectra of GO and MGO samples; Figure S3: Raman spectra of GO and MGO samples; Figure S4: Tensile properties of epoxy and its composites with different MGO: stress-strain curves (a), tensile strength (b), elongation at break (c), and elastic modulus (d).

Author Contributions: X.L.: Conceptualization, Formal analysis, Writing—Original draft preparation; B.Y.: Data curation, Validation, Investigation; J.C.: Writing—Original draft preparation; D.H.: Writing—Original draft preparation; J.L.: Writing—Review and Editing; D.N.: Supervision, Writing—Review and Editing, Funding acquisition. All authors have read and agreed to the published version of the manuscript.

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