

Research Article

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Effect of water immersion on shear strength of epoxy adhesive filled with graphene nanoplatelets

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Abstract: Adhesive bonds are frequently used in industries such as aerospace, automotive, and civil engineering due to their ability to reduce damage to the adherend and their lightweight. However, their application is restricted by their inadequate durability and reliability in hostile environments. Graphene nanoplatelets (GNPs) are employed to enhance epoxy adhesives in this article. The thick adherend shear test is utilized to examine how the shear properties change with different water ageing times. Before exposure to water ageing conditions, GNP-reinforced adhesives exhibit a 3.51% increase compared with neat epoxy at a GNP content of 0.25 wt%. However, after 56 days of water ageing, the increase in shear strength is found to be 13.79%. This suggests that the well-dispersed GNP can reduce the degradation rate in shear properties by half, from 16.71 to 8.44%, at a GNP content of 0.25 wt%. Additionally, as water ageing time increases, the positive influence of GNP on shear properties becomes more evident. The addition of GNP delays the degradation of shear properties caused by water ageing conditions. The effect of GNP does not improve with higher GNP content. When the GNP contents increase to 1 wt%, the shear strength of the GNP-reinforced adhesive decreases compared to neat epoxy.

Keywords: shear property, graphene nanoplatelets, water ageing conditions, digital image correlation

1 Introduction

Adhesive joints are appealing in numerous applications due to their lightweight, reduced damage to the adherends, and other benefits [1,2]. Epoxy has emerged as the most commonly used structural adhesive for bonding various substrates among all adhesives [3,4]. While achieving high initial bonding strength of epoxy adhesive is relatively simple, sustaining good durability in harsh environments can be challenging [5,6]. Water uptake is the foremost factor contributing to the weakening of adhesive joints [7,8]. High levels of moisture or immersion in water are the primary causes of bonding failure during service conditions [9]. The water content in adhesive can significantly alter its mechanical properties and the adhesion interface between the adherends and adhesive [10]. It is almost impossible to keep water from adhesive joints exposed to real working conditions [11]. Water can easily pass through the adherend, adhesive, or migrate along the interface [12]. In the adhesive, cracks and fissures allow water to migrate through capillary action, leading to the irreversible degradation of the epoxy adhesive [13]. In conditions of high moisture, water molecules can induce plasticization and hydrolysis of the adhesive [14]. This can cause a break in the secondary bonds between the adherend/adhesive interface and corrosion of the adherend surface. Ultimately this leads to a noticeable degradation of the mechanical properties of the lap joints [15,16]. Particularly, in the case of metallic bonding, polar bonding surfaces may attract water molecules and weaken the van der Waals force at the interface [17]. After 60 cycles of hydrothermal ageing, the failure strength of single lap joints, scarf joints, and butt joints between aluminium alloy and a polyurethane adhesive decreased in a cubic polynomial pattern with ageing cycles. The initial strength decreased by approximately 30% [18]. In high levels of moisture, the location of failure shifts from cohesive to interface failure [18]. This phenomenon is more evident when water molecules migrate adhesive joints [19]. The lap shear strength of adhesive joints decreased after

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exposure to both distilled water and seawater. The degradation in the adhesive joint strength was particularly pronounced when immersed in distilled water, where a higher moisture ingress into the adhesive occurred [19].

Fillers have been added to the adhesive to increase its durability [20,21], as they were introduced into the free volume of the matrix to reduce polymer chain mobility and delay the plasticization of the adhesive. The addition of 50 wt% aluminum filler in the adhesive does not significantly affect its behaviour when immersed in water [20]. The polymerizable surfactants were investigated to improve the water resistance of pressure-sensitive adhesives [22]. The findings indicate that the use of polymerizable surfactants can improve the adhesive's glass transition temperature, water resistance, shear strength, and deformation energy [22]. Nanofillers with high aspect ratio, like Fullerene nanoparticles, carbon nanotubes (CNT), and graphene nanoplatelets (GNPs), have been preferable for modifying adhesives, reducing their ability to absorb water and enhancing their resistance to hydrothermal degradation [23]. To investigate their effects on water uptake, glass transition temperature, and single lap shear properties of the adhesive film, 0.1 wt% CNT and 5 wt% GNP were added separately to the film [19]. The research results indicated that CNT samples had the highest diffusion coefficient, while GNP specimens had the lowest. Additionally, the glass transition temperature and mechanical properties showed the opposite effect, with CNT samples exhibiting less degradation over time [19]. However, the article did not discuss whether this effect was dependent on the CNT or GNP contents. A mixture of nanofillers can also have a positive effect on resisting hydrothermal ageing. The addition of a mixture of SWCNT and Fullerene nanoparticles reduced the degradation of the adhesive during hydrothermal ageing by limiting the molecular chains of the polymer [23]. Compared to other nanofillers, GNPs possess superior barrier properties in reducing water diffusion into adhesive joints due to their 2D nature and significant aspect ratio. The GNPs induce higher tortuous paths for water diffusion into the adhesive [19]. Further research is necessary to determine the impact of GNPs on the shear properties of epoxy adhesive when exposed to water ageing conditions. Previous studies have shown that moisture has a greater effect on shear properties than on tensile strength [24].

The shear properties of adhesives are typically the primary factor when selecting adhesives, both in research labs during development and in the field during service [25–27]. Information about the impact on the shear properties of epoxy adhesives at various water ageing periods due to the inclusion of GNPs is scarce.

Previously, we tested the mode I fracture toughness of epoxy adhesive reinforced with different amounts of GNP (0.25, 0.5, 0.75, and 1 wt%) under water immersion conditions [6]. After 56 days of immersion, the degradation rate for the 0.25 wt% GNP-reinforced adhesive was the lowest. Further investigation is needed to determine the optimum GNP contents in improving shear strength under water immersion conditions. In this article, different amounts of GNP (0.25, 0.5, 0.75, and 1 wt%) were dispersed in epoxy joints for this study. The shear properties of GNP-reinforced adhesive were then compared to that of neat epoxy under water immersion conditions.

2 Experiment

2.1 Materials

The two-part epoxy adhesive used in this study was mixed in a 3:1 weight ratio using a planetary vacuum mixer (ZYMC-200V, Shenzhen, China). This adhesive was supplied by Shanghai Kangda Chemical and New Material Company, and its cure scheme was 96 h at 25°C. Different weight fractions of GNPs (0.25, 0.5, 0.75, and 1 wt%) were prepared to reinforce the pure epoxy adhesive, GNP was purchased from Kelude Company, and GNP/epoxy composites can be found in previous works [28]. The process involved adding GNPs with controlled weight to acetone at a concentration of $2\text{ mg}\cdot\text{mL}^{-1}$, followed by 6 h of ultrasonication to fully exfoliate and disperse the GNPs into the solvent. To create the composite adhesive, epoxy resin was added to the GNPs/acetone mixture. The mixture was pre-mixed using a magnetic stirrer at room temperature for 3 h. The temperature was then raised to 100°C to completely evaporate the acetone. The mixture of GNPs and epoxy resin was cooled to room temperature before adding the curing agent at a weight ratio of 3:1.

2.2 Thick adherend shear test (TAST) specimen

The shear properties of the GNP-reinforced adhesive were investigated under water ageing conditions using TAST specimens. The dimensions of the TAST specimens were determined based on ASTM D5656-10 and ISO 11003-2, as indicated in Figure 1. Stainless steel with Young's modulus of 210 GPa was chosen as the adherends. To regulate the

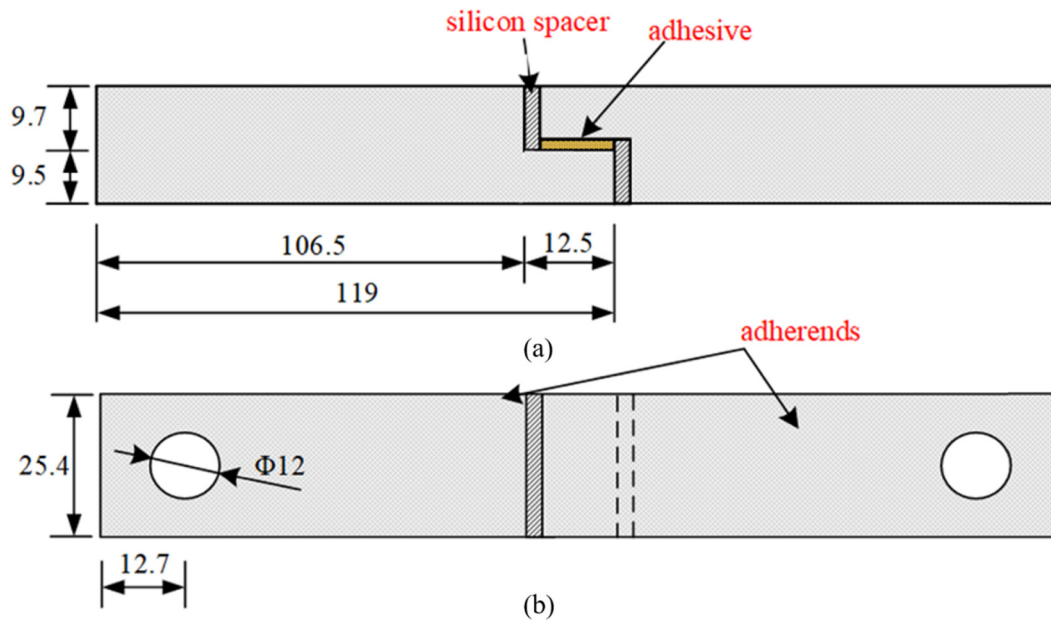


Figure 1: Dimensions of TAST specimens. (a) from side view; (b) from top view (unit: mm).

bond line length of 9.5 mm, 3 mm wide silicone spacers were placed between the two adherends, and the adhesive thickness was designed to be 0.2 mm. The surfaces of all adherends, except the bond surface, underwent anticorrosion treatment to prevent degradation during water immersion.

Prior to adhesive application, the surfaces to be bonded were first cleansed with acetone to remove metal oxides and rust. Afterwards, sanding was done using 24# sandpaper in the $\pm 45^\circ$ direction for 20 min. An electrical sanding machine

was then applied for 5 min in the same direction to further increase surface roughness and ensure good contact between the adhesive and adherends. Finally, the bonded surface was rinsed with acetone to remove any sand grit or dust residue. The side surfaces of the adherends were wiped with the release agent for convenient removal of the excess adhesive. A metal mould was constructed to align the TAST specimens, as illustrated in Figure 2. Before applying the adhesive, the average thickness and width of the overlap region were gauged and documented using a Vernier caliper. After the preparation of TAST specimens, the total thickness of the TAST specimen at the overlap was measured, and the average thickness of the overlap of adherends was subtracted to determine the actual average thickness of the adhesive. Five specimens were fabricated in one batch.

To simulate the water ageing condition, all TAST specimens were immersed in distilled water at a constant temperature of 23°C for 14, 28, 42, and 56 days, respectively, as illustrated in Figure 3, to study the effect of different water ageing time on the shear properties of the epoxy adhesive.

2.3 Testing procedure

The tensile testing for TAST specimens was performed on an electronic universal testing machine (Wance ETM 504C) at a constant crosshead speed of $0.2 \text{ mm} \cdot \text{min}^{-1}$, as shown in Figure 4. Two clamps with a U shape were fabricated to fix the TAST specimens and at least five specimens were tested

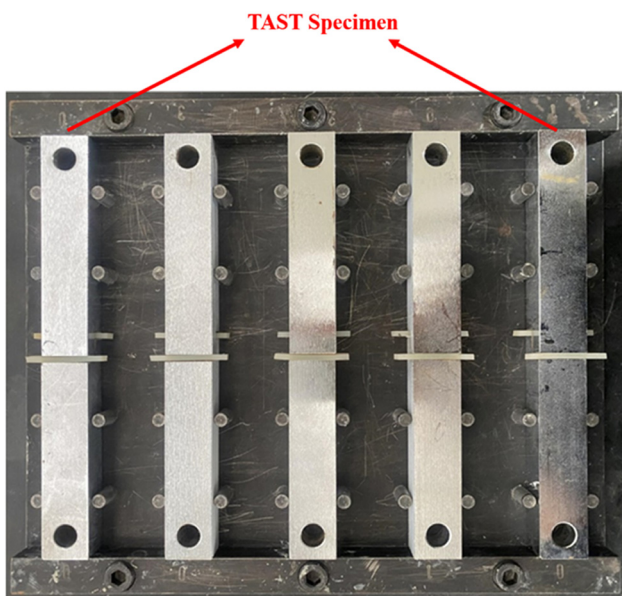


Figure 2: Metal mould for preparation of TAST specimens.



Figure 3: TAST specimens immersed in distilled water.

for repeatability. The force–time curves were recorded during the test.

In the present study, digital image correlation (DIC) technology is used to measure the shear strain of adhesive in the TAST specimen. DIC technology is now widely used in various fields to analyze full-field displacement changes of the specimen by recording its relative movement in real time [29,30]. This method minimizes the deformation of adherends and clamps used to fix the TAST specimens. The shear strain of the adhesive can be obtained using DIC technology, rather than the shear strain of the entire TAST specimen [29]. To aid in tracking the displacement of

the adhesive, uniform speckles were sprayed onto the TAST specimens, as shown in Figure 5, and the local overlap region was the focus during testing. The shear strain of the adhesive was measured and analyzed using the GOM ARAMIS 12 M system and high-resolution cameras. The picture capture rate was set to be 2 Hz for the entire test, and the reference stage was defined prior to the experiment.

According to the data process provided by Kosmann *et al.* [29], the shear strain of the adhesive could be obtained by DIC technology. First, the origin and coordinate axis of the system is defined, as shown in Figure 6. In this article, the y-axis is parallel to the bond line and x-axis is perpendicular to the y axis; z axis is out of the plane. Second, the actual thickness of the adhesive was used to define the scale and calibrate this 2D measurement system. Calibration is necessary for this 2D system as it was not calibrated using the calibration target that is always available in the 3D measurement system. In the calculation of the shear strain of adhesive, to avoid the deformation of adherends, several points were defined as follows: Point_1 is located in the middle of the adhesive, while Point_3 and Point_4 are positioned 10 mm away from Point_1. To ensure accuracy, we created two extensometers, Extensometer_1 and Extensometer_3, both parallel to the x-axis and 10 mm in length. Point_3 and Point_4 are extracted at the end of each extensometer. Point_2 and Point_5 are another 10 mm away from Point_3 and Point_4, shown in Figure 6. Two additional extensometers have been defined to measure these points, as in the previous case.

Based on the methods provided by Korsman, the displacement information for each point in every direction is

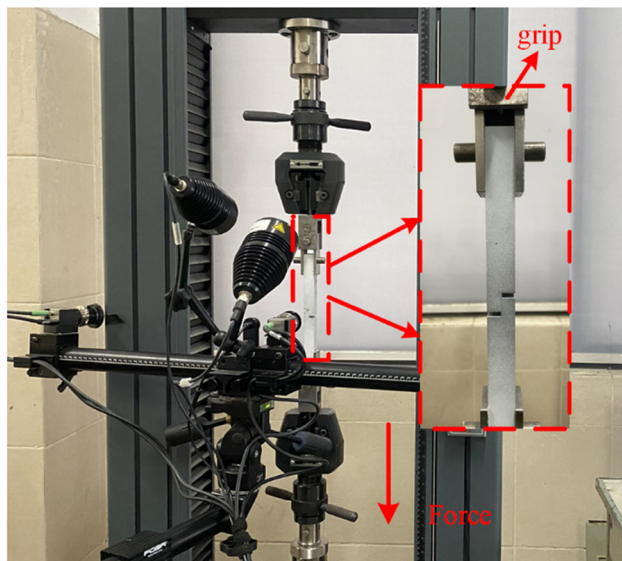


Figure 4: Testing procedure for TAST specimens.

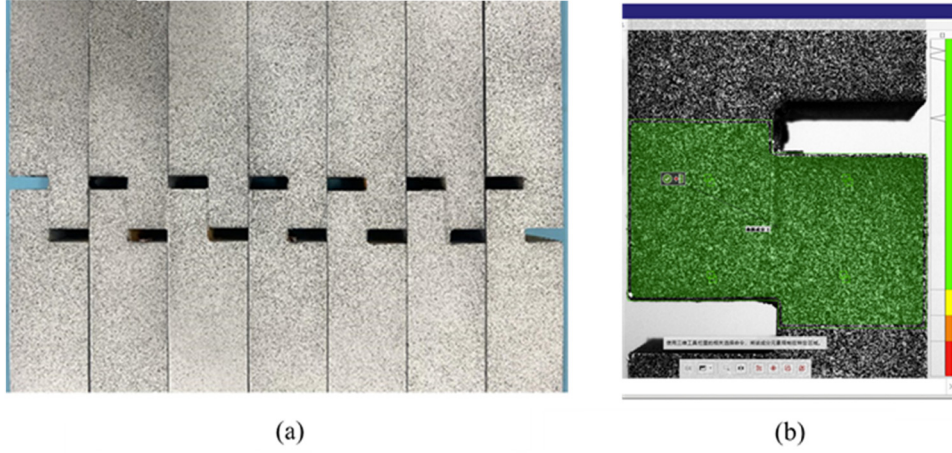


Figure 5: Preparation for DIC test of TAST specimens. (a) TAST specimens with speckles and (b) TAST specimens in the ARAMIS system.

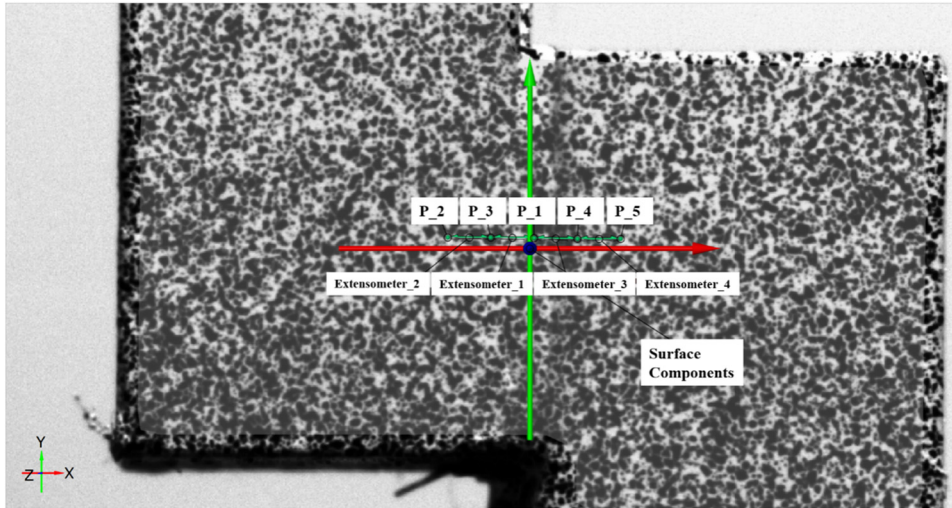


Figure 6: Points distribution and data evaluation of DIC test.

exported for each image. This approach eliminates the need for reference specimens to correct adherend deformation. The shear strain of the adherends is calculated using Eqs. (1) and (2). The relative movement between Point_2 and Point_3, Point_4 and Point_5, respect to their normalized distance $\text{Dist}_{P_2P_3}$ and $\text{Dist}_{P_4P_5}$, are used to calculate the shear strain of adherend. The shear strain of the adhesive is determined by analyzing the deformation data of Point_3 and Point_4 to correct the deformation of

adherends, as shown in Eq. (3). The shear strength of the adhesive was obtained by Eq. (4). The shear strain-strength of the adhesive was obtained from the data collected by the universal testing machine and DIC.

$$\gamma_{adh1} = \frac{P2y - P3y}{\text{Dist}_{P2P3}}, \quad (1)$$

$$\gamma_{adh2} = \frac{P4y - P5y}{\text{Dist}_{P4P5}}, \quad (2)$$

$$\gamma_{adhesive} = \frac{(P3y - P4y) - \gamma_{adh1} \times 0.5 \times (\text{Dist}_{P3P4} - T) - \gamma_{adh2} \times 0.5 \times (\text{Dist}_{P3P4} - T)}{T}, \quad (3)$$

$$\tau = \frac{P}{B \times L}, \quad (4)$$

where γ_{adh1} and γ_{adh2} represent the shear strain of two adherends, $\gamma_{adhesive}$ is the shear strain of adhesive; τ is the shear strength of adhesive, P represents the force value obtained from the universal testing machine; B and L are the overlap width and length respectively; T is the thickness of adhesive; $Dist_{P3P4}$ is the distance between Point_3 and Point_4; $P2y$ is the displacement of Point_2 in the y -direction, and these data are obtained from the DIC test; $P3y$ is the displacement of Point_3 in the y -direction, *etc.*

3 Results and discussion

3.1 Shear properties of GNP- reinforced epoxy adhesive before water ageing conditions

The shear stress–strain curves were obtained for GNP-reinforced adhesive with varying GNP contents, as shown in

Figure 7. Shear strength, failure shear strain, and shear modulus were then calculated. Shear modulus was determined as the initial slope of the shear stress–strain curves within the shear strain range of 0.5–1%. When compared to neat epoxy adhesive before water ageing conditions, the shear strength of GNP-reinforced adhesive displays an increasing trend, but not significantly, with the most considerable improvement being 4.7% when the GNP content is 0.5 wt%. The increase in failure shear strain becomes more noticeable when adding GNP to the adhesive, resulting in a 23% increase with 0.25 wt% GNP content. The well-dispersed two-dimensional GNP may have increased the tortuosity of crack propagation, reducing the speed of crack propagation. This, in turn, could have increased the failure shear strain of the GNP-reinforced adhesive. However, the incorporation of GNP had little positive effect on the shear modulus of epoxy adhesive, the shear modulus is essentially unchanged or even reduced by the addition of the GNP. Increasing the GNP content did not result in any further improvement in the shear properties, and in fact showed a decreasing trend in shear strain when the GNP content was 1 wt%.

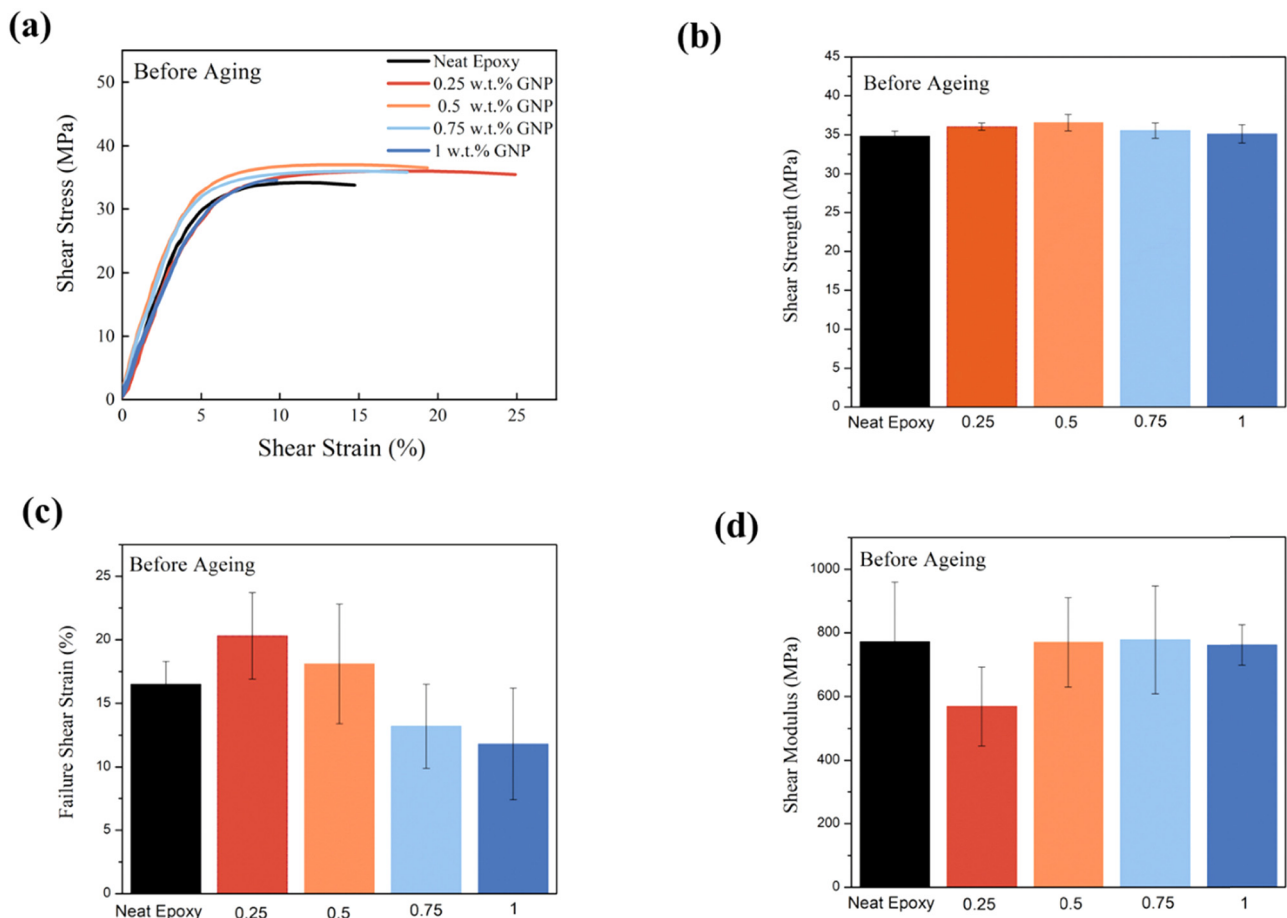


Figure 7: Shear properties of GNP-reinforced adhesive with varying GNP contents before water ageing conditions: (a) typical shear stress–strain curves, (b) shear strength, (c) failure shear strain, and (d) shear modulus.

3.2 Effect of water ageing conditions on shear properties of GNP-reinforced adhesive

To investigate the effect of GNP reinforcement on the shear properties of epoxy adhesive under water ageing conditions, the ageing time was varied and the resulting changes in shear properties were observed.

3.2.1 After 14 days water ageing conditions

During the 14 days ageing conditions, GNP incorporation did not result in much difference in shear strength and shear modulus between neat epoxy and GNP-reinforced adhesive, as shown in Figure 8. However, the incorporation of 0.25 wt% GNP resulted in a significant 38% growth in failure shear strain. Higher GNP content still leads to a decreasing trend in the shear properties of epoxy adhesive after 14 days of water ageing. The effective dispersion of

GNP may block water molecule diffusion, reducing plasticity caused by water and increasing the modulus of the epoxy adhesive.

3.2.2 After 28 days water ageing conditions

Similar to previous results, under 28-day ageing conditions, the addition of GNP did not yield a significant increase in shear strength and shear modulus, even resulted in a declining trend in shear modulus when GNP was added to the neat epoxy adhesive, as illustrated in Figure 9. The behaviour of properties in a thin layer of adhesive may differ from the bulk properties of the adhesive. The increase in shear modulus resulting from the addition of GNP was not as significant as the increase in tensile modulus [1]. Incorporating GNP largely boosted the failure shear strain of the epoxy adhesive, particularly at a GNP content of 0.25 wt% wherein an increase of 37% was observed compared to the neat epoxy adhesive.

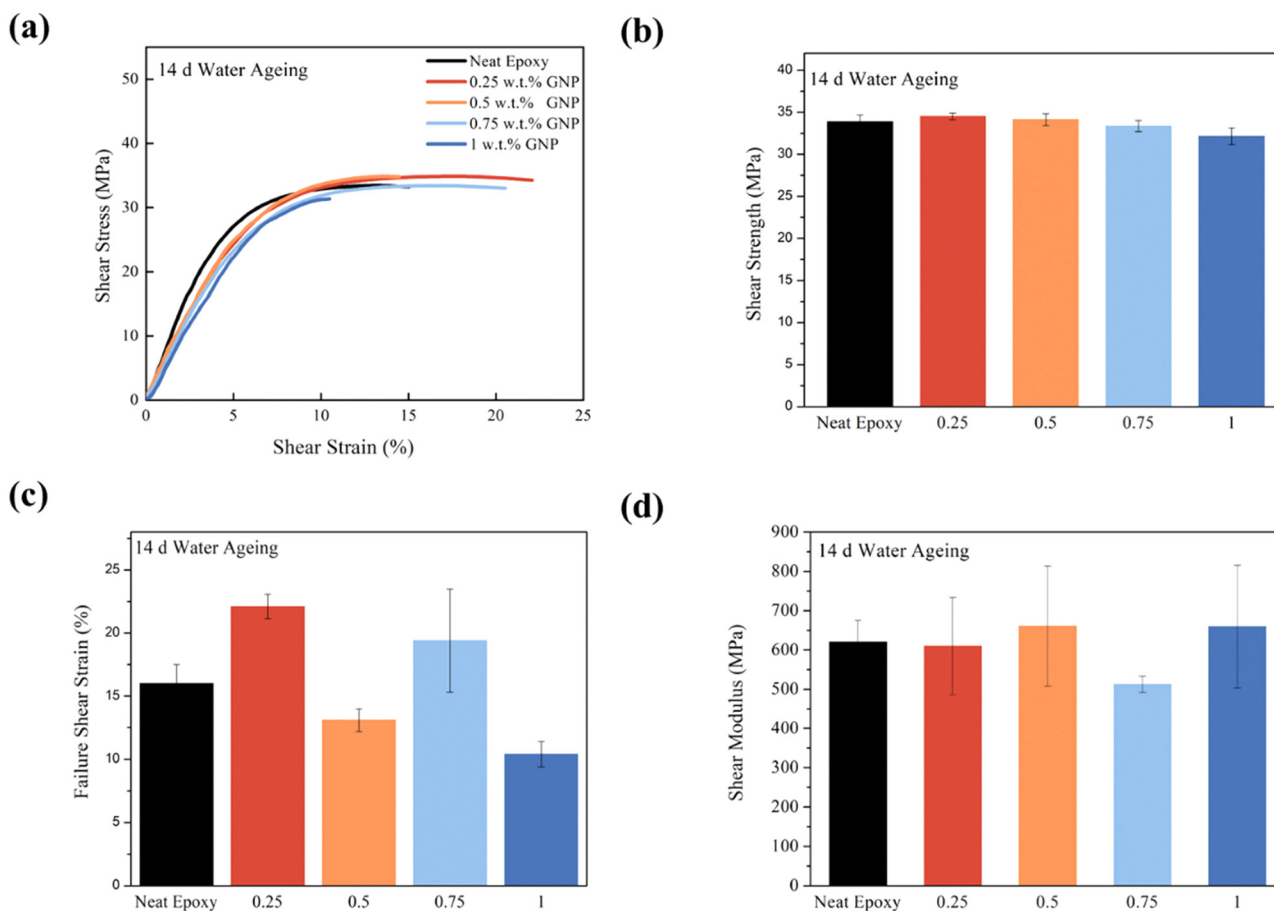


Figure 8: Shear properties of GNP-reinforced adhesive with varying GNP contents after 14 days of water ageing conditions: (a) typical shear stress-strain curves, (b) shear strength, (c) failure shear strain, and (d) shear modulus.

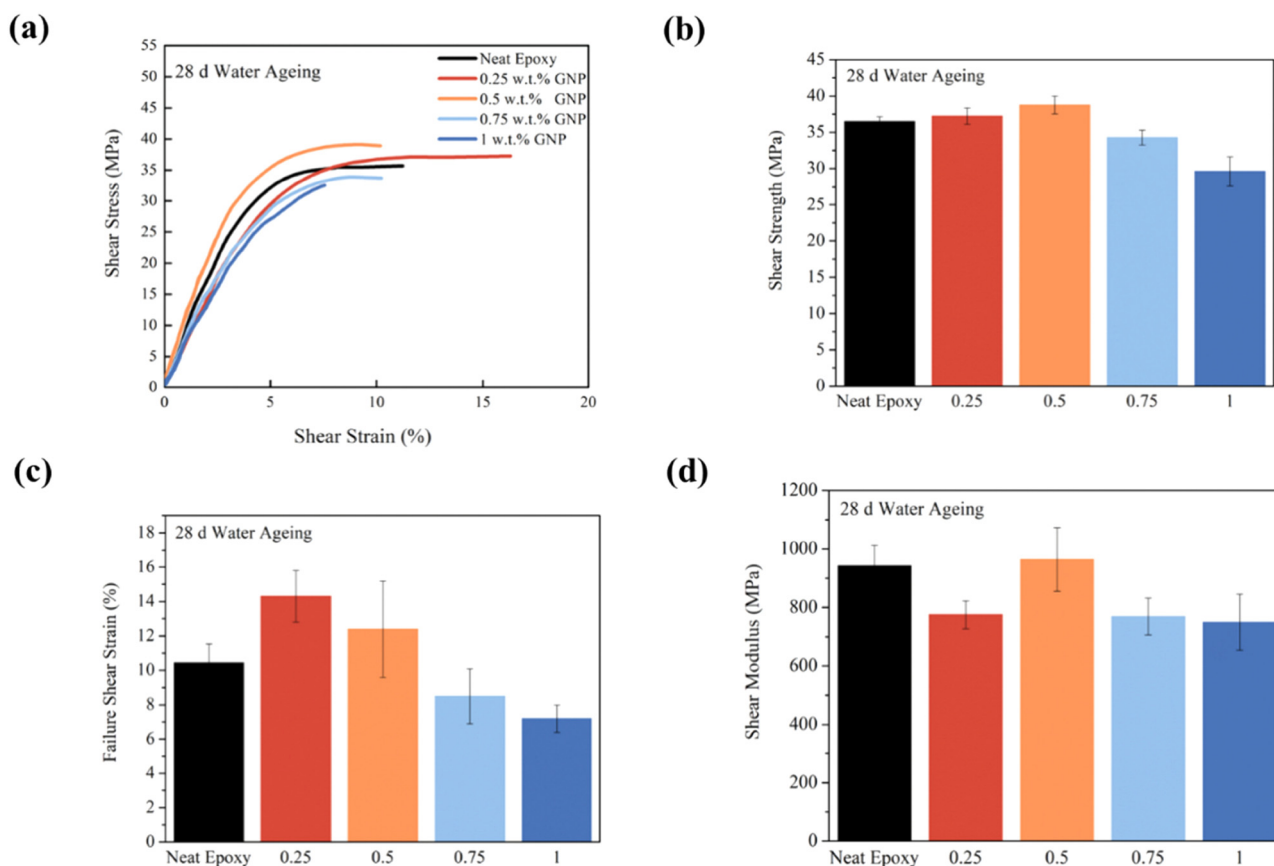


Figure 9: Shear properties of GNP-reinforced adhesive with varying GNP contents after 28 days of water ageing conditions: (a) typical shear stress-strain curves, (b) shear strength, (c) failure shear strain, and (d) shear modulus.

Interestingly, as the water ageing time extended to 28 days, both the shear strength and shear modulus of the neat epoxy and GNP-reinforced adhesive increased compared to non-ageing and 14 days water ageing conditions. Nevertheless, the adhesive reinforced with higher GNP content, 0.75 and 1 wt%, displayed a decreasing trend. The reason why the shear strength of adhesive increases may be due to the post-cure effect of adhesive outweighing the negative impact of water ageing. However, a higher GNP content in the epoxy adhesive could cause aggregation, according to our previous work on scanning electron microscope of neat epoxy adhesive and GNP-reinforced adhesive [1], agglomeration was observed in the 0.75 wt% GNP-reinforced adhesive which may account for the decreased trend in shear properties of epoxy adhesive under water immersion conditions.

3.2.3 After 42 days water ageing conditions

After prolonging the water ageing time to 42 days, the shear strength difference between the neat epoxy adhesive

and GNP-reinforced adhesive became more apparent, with an increase of 12.7% when the GNP content was 0.5 wt%, as shown in Figure 10. An evident rise in the failure shear strain was also noticed in an adhesive reinforced with 0.5 wt% GNP. This reinforcement allows the adhesive to deform more before failure. The shear modulus of GNP-reinforced adhesive increased obviously compared with neat epoxy, with the 1 wt% GNP-reinforced adhesive increased by 33% and 0.75 wt% GNP-reinforced adhesive increased by 18%. This rise in shear modulus may be attributed to adhesive hardening resulting from the post-curing effect in these two adhesives with greater GNP reinforcement.

Compared to the 28 days water ageing conditions, the shear strength and shear modulus of neat epoxy decreased while the epoxy adhesive reinforced with varying GNP content exhibited divergent trends. Specifically, with lower GNP content of 0.25 and 0.5 wt%, the shear properties remained largely unchanged upon the extension of ageing time to 42 days. On the other hand, with GNP content of 0.75 and 1 wt%, there was a marked increase in the shear strength. The results suggest that higher GNP content

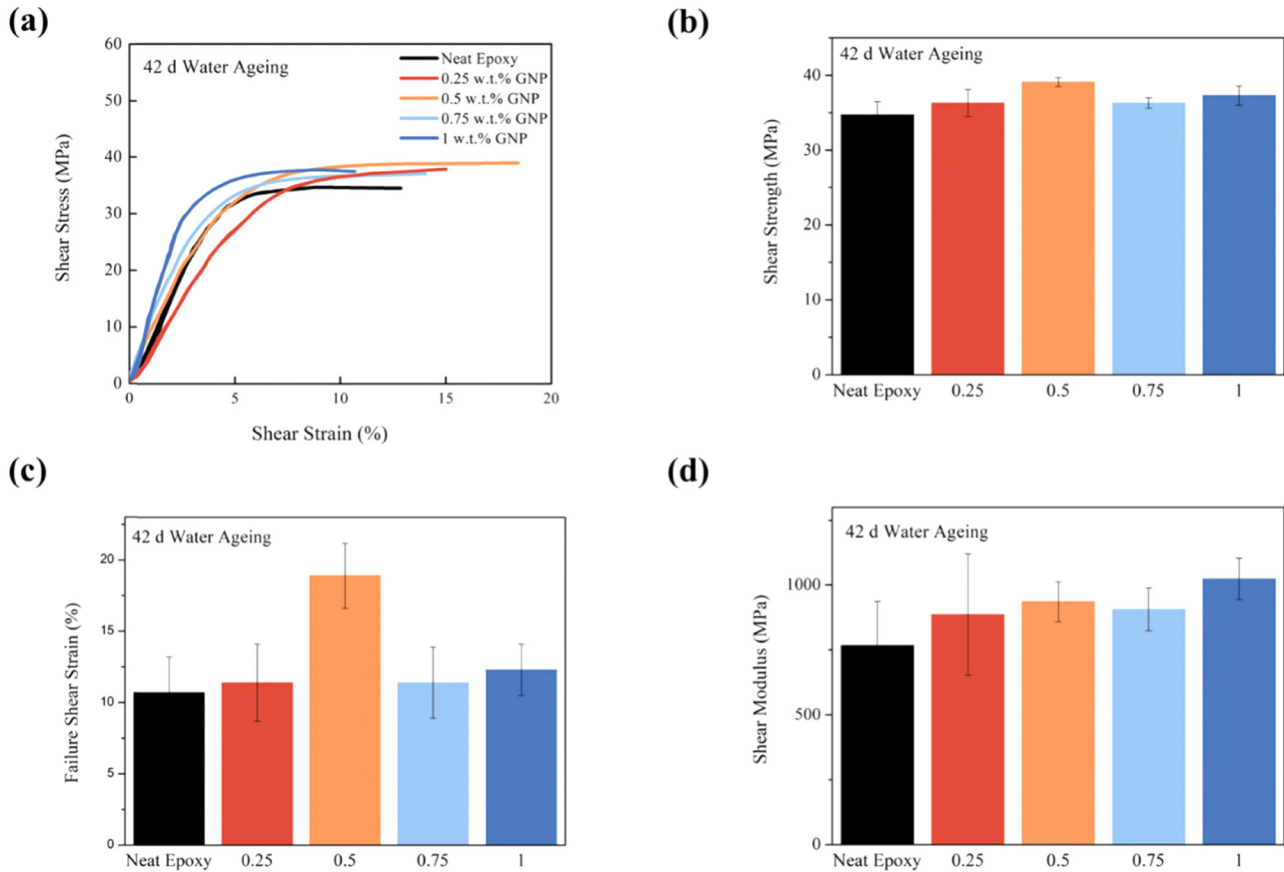


Figure 10: Shear properties of GNP-reinforced adhesive with varying GNP contents after 42 days of water ageing conditions: (a) typical shear stress–strain curves, (b) shear strength, (c) failure shear strain, and (d) shear modulus.

delays the epoxy hardening effect of the post-cure process. Conversely, adding less GNP balances the positive post-cure effect and the negative effect of water ageing. However, the negative effect overtakes the post-cure effect in neat epoxy at longer water ageing conditions, resulting in irreversible degradation.

3.2.4 After 56 days of water ageing conditions

Under the 56 days water ageing conditions, the disparity between neat epoxy and GNP-reinforced adhesive was the highest in comparison to the previous ageing conditions (Figure 11). When GNP content was 0.25 wt%, the shear strength increased by 13.79% compared to the neat epoxy. However, before the ageing process, the increase was only 3.51%. With extended water ageing conditions, the shear strength increase was more apparent with the addition of 0.25 wt% GNP. In contrast to non-ageing conditions, neat epoxy experienced apparent shear strength degradation of 16.71% after 56 days of water ageing, while the 0.25 wt% GNP-reinforced adhesive decreased by 8.44%. The property

degradation magnitude of 0.25 wt% GNP-reinforced adhesive was significantly smaller than that of neat epoxy. Based on our previous study on the distribution of moisture concentration in a 0.25 wt% GNP-reinforced epoxy adhesive layer after 56 days of water immersion conditions [6], we found that the moisture diffusion constants in the 0.25 wt% GNP-reinforced adhesive were lower. As a result, the moisture content of the 0.25 wt% GNP-reinforced adhesive decreased by 19.3% compared to the neat epoxy after 56 days of water immersion. That is the main reason for less degradation in shear properties under water immersion conditions. When a higher GNP content is utilized to reinforce the neat epoxy, it notably shows a decreasing trend when the GNP content is at 1 wt%.

Similar to the trend in shear strength, the shear modulus of 0.25 wt% GNP reinforced adhesive showed a significant increase of 53.43% compared to neat epoxy after 56 days of water ageing. Compared to the non-ageing conditions, the shear modulus of the neat epoxy decreased by 16.84%. Interestingly, the shear modulus of the adhesive with 0.25 wt% GNP-reinforced adhesive displayed an increasing trend after 56 days of water ageing conditions. The addition of dispersed GNP content may have improved

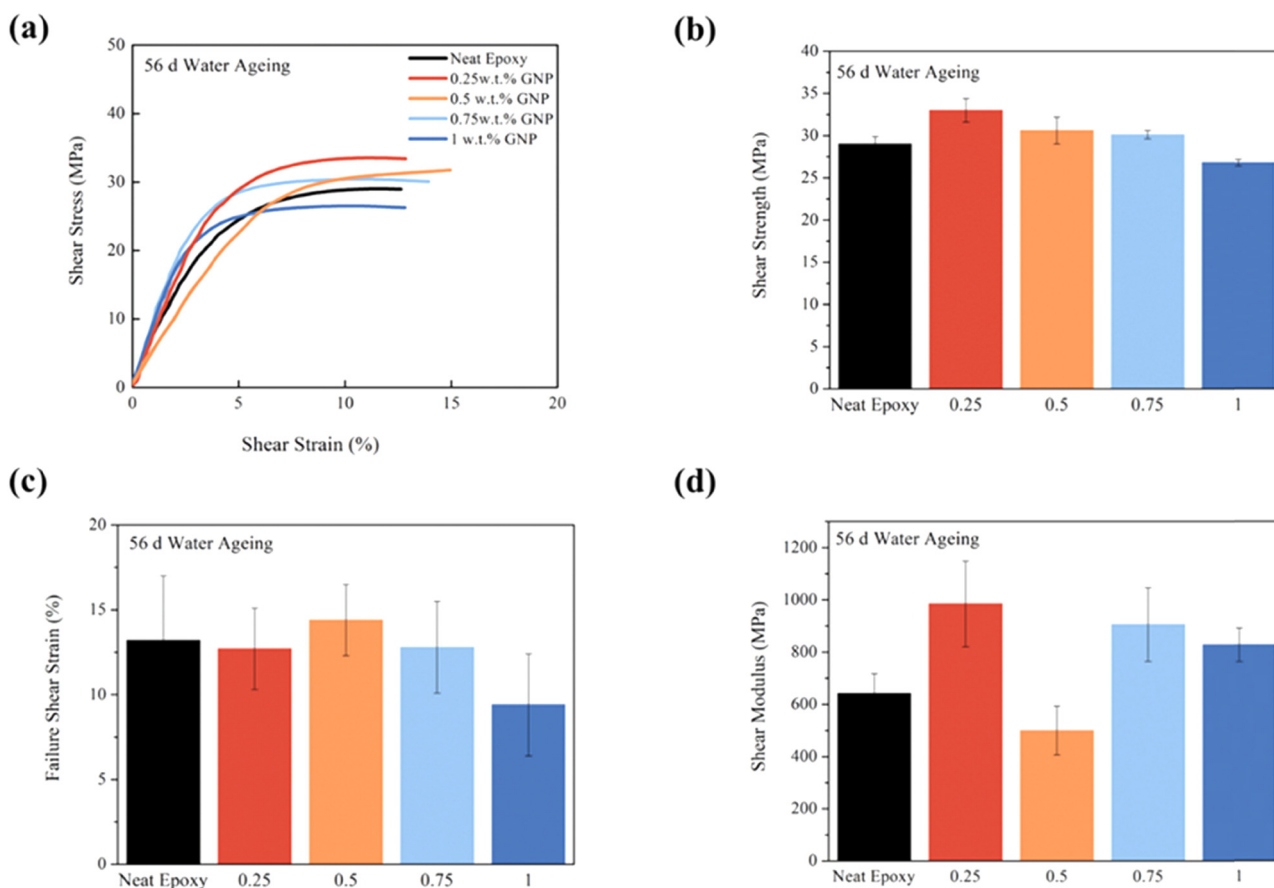


Figure 11: Shear properties of GNP-reinforced adhesive with varying GNP contents after 56 days of water ageing conditions: (a) typical shear stress-strain curves, (b) shear strength, (c) failure shear strain, and (d) shear modulus.

the post-cure process, reduced the mobility of epoxy molecule chains, and prevented water molecules from causing plasticity. Regarding failure shear strain, GNP-reinforced adhesive exhibited no significant increase in comparison to neat epoxy. The magnitude of the increase was even smaller than that of non-ageing conditions. This suggests that GNP addition mainly increased the shear modulus and shear strength during longer water ageing conditions, as opposed to failure shear strain, which indicates the adhesive's deformation capability.

To summarize, during the design stage of reducing shear property degradation under water immersion conditions, it was suggested to add 0.25 wt% GNP to the epoxy adhesive to enhance failure shear strain by 37% for immersion periods less than 28 days. However, for longer immersion periods (56 days), the addition of 0.25 wt% GNP-reinforced adhesive increased the shear strength and shear modulus by 13.79 and 53.43%, respectively.

4 Conclusions

The present study analyzed the shear properties of GNP-reinforced epoxy adhesive under water ageing conditions, including shear strength, failure shear strain and shear modulus. DIC technology was employed to measure the shear strain of the adhesive. Various levels of GNP content (0.25, 0.5, 0.75, and 1 wt%) were used to modify the neat epoxy, leading to improvement of the adhesive's shear properties under different water ageing periods (14, 28, 42, and 56 days).

Before water ageing conditions, the GNP-reinforced adhesive showed a 4.7% increase in shear strength compared to the neat epoxy. The addition of GNP did not alter the shear modulus, yet it showed a marked improvement in failure shear strain. When immersed in water for short periods (less than 28 days), no obvious difference in shear strength and shear modulus was observed between the

neat adhesive and GNP-reinforced adhesive. However, there was a noticeable increase in failure shear strain, which ranged from 23 to 37%. When the water ageing time was prolonged to 56 days, the difference in shear strength and shear modulus between neat epoxy and GNP reinforced adhesive became the most apparent. The addition of 0.25 wt% GNP resulted in a 13.79% increase in shear strength and a 53.43% increase in shear modulus compared to the neat epoxy.

On the other hand, compared with the non-ageing conditions, both the neat epoxy and GNP-reinforced adhesive initially decreased, then increased, and finally decreased again when immersed in water for varying periods of time. The increased trend in shear strength was probably due to the post-cure of adhesive which outweighed the negative effect of water ageing. The addition of GNP could reduce the negative effects of water ageing on the shear strength degradation of epoxy adhesive. The neat epoxy adhesive showed an apparent shear strength degradation of 16.71% after 56 days of water ageing, while the 0.25 wt% GNP-reinforced adhesive decreased by only 8.44%. The findings demonstrated that effectively dispersed GNP could improve the shear properties of adhesive under water ageing conditions, and this effect became more pronounced as the ageing time was extended. However, using higher GNP contents to reinforce adhesive did not show a more significant increase in shear properties. In fact, there was a decreasing trend observed when the GNP contents increased to 1 wt%. Therefore, it is suggested to add 0.25 wt% GNP to the epoxy adhesive to improve its shear properties under water ageing conditions.

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Data availability statement: The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

References

- [1] Jia, Z., J. Yu, Q. Liu, S. Yu, and Z. Wang. Functionally graded adhesive joints with exceptional strength and toughness by graphene nanoplatelets reinforced epoxy adhesives. *International Journal of Adhesion and Adhesives*, Vol. 125, 2023, id. 103402.
- [2] Jia, Z., G. Yuan, D. Hui, X. Feng, and Y. Zou. Effect of high strain rate and low temperature on mode II fracture toughness of ductile adhesive. *International Journal of Adhesion and Adhesives*, Vol. 86, 2018, pp. 105–112.
- [3] Ranji, S. and M. C. Lee Study on synthesizing new urethane epoxy adhesives and their adhesive properties on different substrates. *International Journal of Adhesion and Adhesives*, Vol. 117, 2022, id. 103174.
- [4] Wang, X., X. Zhang, X. Bian, C. Lu, Q. Li, C. Bai, et al. Curing kinetics and mechanical properties of an underwater SiO₂/epoxy adhesive. *The Journal of Adhesion*, Vol. 99, No. 10, 2022, pp. 1607–1625.
- [5] Elhag, A. B., A. Raza, Q. Z. Khan, M. Abid, B. Masood, M. Arshad, et al. A critical review on mechanical, durability, and microstructural properties of industrial by-product-based geopolymer composites. *Reviews on Advanced Materials Science*, Vol. 62, No. 1, 2023, id. 20220306.
- [6] Liu, Q., M. Tao, J. Yu, Y. Zou, and Z. Jia Effect of graphene nanoplatelets on mode I fracture toughness of epoxy adhesives under water aging conditions. *The Journal of Adhesion*, Vol. 100, No. 4, 2024, pp. 267–289.
- [7] Bruckner, T. M., T. D. Singewald, R. Gruber, L. Hader-Kregl, M. Klotz, M. Müller, et al. Water absorption and leaching of a 1K structural model epoxy adhesive for the automotive industry. *Polymer Testing*, Vol. 117, 2023, id. 107870.
- [8] Shaafaey, M., W. Miao, A. Bahrololoumi, O. Nabinejad, and R. Dargazany. Effects of hydrolytic aging on constitutive behavior of silicone adhesives in seawater and distilled water. *Experimental Mechanics*, Vol. 63, No. 1, 2022, pp. 139–162.
- [9] Shin, P. S., D. J. Kwon, J. H. Kim, S. I. Lee, K. L. DeVries, and J. M. Park. Interfacial properties and water resistance of epoxy and CNT-epoxy adhesives on GFRP composites. *Composites Science and Technology*, Vol. 142, 2017, pp. 98–106.
- [10] Ceylan, R., E. Ozun, O. Çoban, M. Ö. Bora, and T. Kutluk. The effect of hygrothermal aging on the adhesion performance of nanomaterial reinforced aluminium adhesive joints. *International Journal of Adhesion and Adhesives*, Vol. 121, 2023, id. 103304.
- [11] Machalická, K., M. Vokáč, M. Kostecká, and M. Eliášová. Structural behavior of double-lap shear adhesive joints with metal substrates under humid conditions. *International Journal of Mechanics and Materials in Design*, Vol. 15, No. 1, 2018, pp. 61–76.
- [12] Mu, W. L., Q. H. Xu, J. Na, Y. Fan, Y. Sun, and Y. Liu. Investigating the failure behavior of hygrothermally aged adhesively bonded CFRP-aluminum alloy joints using modified Arcan fixture. *Thin-Walled Structures*, Vol. 182, 2023, id. 110303.
- [13] Sousa, J. M., J. R. Correia, and S. Cabral-Fonseca. Some permanent effects of hygrothermal and outdoor ageing on a structural polyurethane adhesive used in civil engineering applications. *International Journal of Adhesion and Adhesives*, Vol. 84, 2018, pp. 406–419.

- [14] Mirzaei, G., B. Mohebbi, and M. Tasooji. The effect of hydrothermal treatment on bond shear strength of beech wood. *European Journal of Wood and Wood Products*, Vol. 70, No. 5, 2012, pp. 705–709.
- [15] Tomita, Y., I. Shohji, S. Koyama, and S. Shimizu. Degradation behaviors of adhesion strength of structural adhesive for weld-bonding under high temperature and humidity conditions. *Procedia Engineering*, Vol. 184, 2017, pp. 231–237.
- [16] Tedesco, T. K., F. B. T. Alves, T. L. Lenzi, A. F. B. Calvo, A. Reis, A. D. Loguerio, et al. Effect of 2 years water aging on bond strength stability of adhesive systems to artificial caries-affected primary dentin. *International Journal of Adhesion and Adhesives*, Vol. 54, 2014, pp. 172–176.
- [17] Pawlik, M., Y. Lu, and H. Le. Effects of surface modification and graphene nanoplatelet reinforcement on adhesive joint of aluminium alloys. *International Journal of Adhesion and Adhesives*, Vol. 99, 2020.
- [18] Qin, G., Y. Fan, J. Na, W. Mu, and W. Tan. Durability of aluminium alloy adhesive joints in cyclic hydrothermal condition for high-speed EMU applications. *International Journal of Adhesion and Adhesives*, Vol. 84, 2018, pp. 153–165.
- [19] Sánchez-Romate, X. F., P. Terán, S. G. Prolongo, M. Sánchez, and A. Ureña. Hydrothermal ageing on self-sensing bonded joints with novel carbon nanomaterial reinforced adhesive films. *Polymer Degradation and Stability*, Vol. 177, 2020, id. 109170.
- [20] Al-Harathi, M., R. Kahraman, B. Yilbas, M. Sunar, and B. J. A. Aleem. Influence of water immersion on the single-lap shear strength of aluminum joints bonded with aluminum-powder-filled epoxy adhesive. *Journal of Adhesion Science and Technology*, Vol. 18, No. 15–16, 2004, pp. 1699–1710.
- [21] Nijemeisland, M., Y. V. Metelewa-Fischer, and S. J. Garcia. Identifying interfacial failure mode in aerospace adhesive bonds by broadband dielectric spectroscopy. *International Journal of Adhesion and Adhesives*, Vol. 118, 2022, id. 103246.
- [22] Márquez, I., N. Paredes, F. Alarcia, and J. I. Velasco. Influence of polymerizable surfactants on the adhesion performance and water resistance of water-based acrylic pressure-sensitive adhesives (PSAs). *Journal of Adhesion Science and Technology*, Vol. 37, No. 11, 2022, pp. 1770–1788.
- [23] Esmail, R., M. H. Alaei, J. Eskandarijam, and M. Rezvaninasab. Effect of presence of SWCNT and Fullerene nano particles in the adhesive of CFRP single lap joint on the lap shear strength subjected to hygrothermal conditions. *Journal of Adhesion Science and Technology*, Vol. 37, No. 21, 2023, pp. 3013–3029.
- [24] Zhang, J., X. Cheng, X. Guo, J. Bao, and W. Huang. Effect of environment conditions on adhesive properties and material selection in composite bonded joints. *International Journal of Adhesion and Adhesives*, Vol. 96, 2020, id. 102302.
- [25] da Silva, L. F. M., R. A. M. da Silva, J. A. G. Chousala, and A. M. G. Pintob. Alternative methods to measure the adhesive shear displacement in the thick adherend shear test. *Journal of Adhesion Science and Technology*, Vol. 22, No. 1, 2012, pp. 15–29.
- [26] Jia, Z., G. Yuan, X. Feng, Y. Zou, and J. Yu. Shear properties of polyurethane ductile adhesive at low temperatures under high strain rate conditions. *Composites Part B: Engineering*, Vol. 156, 2019, pp. 292–302.
- [27] Kadioglu, F., L. F. Vaughn, F. J. Guild, and R. D. Adams. Use of the thick adherend shear test for shear stress–strain measurements of stiff and flexible adhesives. *The Journal of Adhesion*, Vol. 78, No. 5, 2002, pp. 355–381.
- [28] Wang, Z., Z. Jia, X. Feng, and Y. Zou. Graphene nanoplatelets/epoxy composites with excellent shear properties for construction adhesives. *Composites Part B: Engineering*, Vol. 152, 2018, pp. 311–315.
- [29] Kosmann, J., O. Völkerink, M. J. Schollerer, D. Holzhüter, and C. Hühne. Digital image correlation strain measurement of thick adherend shear test specimen joined with an epoxy film adhesive. *International Journal of Adhesion and Adhesives*, Vol. 90, 2019, pp. 32–37.
- [30] Curt J., M. Capaldo, F. Hild, and S. Roux. Optimal digital color image correlation. *Optics and Lasers in Engineering*, Vol. 127, 2020, id. 105896.