International Research Journal of Advanced Engineering and Science
ISSN (Online): 2455-9024

Investigation the Effect of Clay Nanoparticles on Shear Strength of Adhesively Bonded Composites

Bashar R. AL-Ogaidi
Energy and Renewable Energies Technology Center, University of Technology-Iraq, Baghdad, Iraq
Email address: 130110[AT]uotechnology[DOT]edu[DOT]iq

Abstract— *An adhesive bonding can be ranged among technologies of materials bonding which are used in all industrial branches Adding particles to the adhesive layer in adhesively bonded joints can increase adhesive toughness and improve stress distribution in the adhesive layer. In this investigation, the influence of clay nanoparticles on the adhesion between epoxy adhesive and composite substrates was primarily investigated. Shearing characteristics of modified epoxy were studied by single lap joints. The adhesives were produced from different amounts of Nano clay particles incorporated into epoxy. Glass fiber composite plates were chosen as adherends. The results of shear tests indicated that clay nanoparticles had great influence on adhesion strength. Compared to neat epoxy, it was found that the adhesive strength is increased considerably under shear loadings.*

Keywords— *Adhesive, bonded joints, Nano clay, shear, epoxy.*

I. Introduction

Over the years, metal-based engineering materials have been increasingly replaced by the polymer composites because of advantageous features of polymers including high strength-to-weight ratio, excellent corrosion resistance, superior thermal insulation, and low thermal expansion coefficient. The fiber composite approach can provide significant improvements in specific (property/density) strength and stiffness over conventional metal alloys. Light weight composite materials such as carbon fiber reinforced epoxy resins and glass fiber reinforced laminates are increasingly finding application in aircraft to replace traditional heavier metal structures [1,2].

Recent advances in composites manufacturing technologies provide affordable solutions to the production of complex and large composite parts. To join such composite parts, polymer adhesives are commonly used. Using adhesive bonding for joining composite parts provides many advantages such as low cost, high strength to weight ratio, low stress concentration, fewer processing requirements and superior fatigue resistance and environmental resistance [3,4,5]. Adhesive bonding is rapidly developing for many applications in: aviation, electrical, automotive and marine industries, building engineering, medicine (dentistry). Increases in specific strength, corrosion resistance plus possibility of joining different materials are the principal advantages of adhesive joints over the 'traditional' joining methods (riveting, bolting, welding, brazing). Naturally there is still need to increase mechanical properties and durability of adhesives [1,4,6-8]. The most popular adhesives with high cohesive performance used in structural applications are epoxies, which promise strong, resistant and durable joints. A number of techniques have been considered to improve the mechanical properties of structural adhesives containing fillers such as carbon, nylon, or glass micro- or Nano-fibers and nanographene [7-11]. From a general point of view, nanostructured materials are expected to yield improvements over neat polymers in a wide range of properties, such as mechanical properties [9,10,11,12,13], dimensional stability and barrier properties [14], thermal degradation resistance [15,16], etc.

The Nano clay particles are formed from clay platelets which are stacked together. The thickness and aspect ratio of the clay platelet are about 1 nm and 100, respectively [17]. Due to the large surface area of the Nano-sized particles only small amounts are needed to cause significant changes in the resulting properties of the Nano-composite adhesives [18]. One possibility to increase adhesives mechanical properties is to reinforce them with nanoparticles and nanotechnology [4,19-21]. Adding alumina Nano-fibers to epoxy adhesives slightly increases the strength of sample aluminum joints [19,20]; whereas, the effect of Nano-reinforced composites on the toughness of carbon/epoxy composite joint is significant, vary based on the fabrication method (prefabricated versus co-cured). Previous studies [22,23-36] conducted several studies into the effect of adding alumina nanoparticles to epoxy resin for bonding steel samples. In prior work [37-40] developed epoxy adhesives reinforced with carbon nanotubes, to be utilized in aluminium joints. In particular, they reported that the addition of carbon nanotubes in concentrations up 1 wt% greatly improved the durability of epoxy-based adhesive joint in tests under water at 60◦C. Recently, [28] analyzed the electrical conductivity and the shear strength of polyurethane adhesives filled with different kinds of modified graphite, finding that the strength of the adhesive joints to aluminum increased up to a filler content of 20 wt. %. Patel et al. [27] synthesized Nano-composite adhesives based on acrylic polymers and silica or clay, in order to investigate the effect of these nanoparticles on the adhesion behavior of the hybrid adhesives against different substrates (aluminium, wood, polypropylene). Aluminium and wood joints displayed higher joint strength, because of the interaction of the adhesive with the hydroxyl groups present on the surface of these substrates. Guadagno et al. [17] have investigated the mechanical performance and morphology of structural adhesives modified by graphene additive. They have shown that considerable enhancement was achieved in epoxy adhesives filled with 1 wt% graphene due to the cumulative influence of inter-molecular interactions among the graphene particles and epoxy adhesive [41-46]. Earlier study [47] studied the effect of adding graphene on low viscosity adhesives and have shown that the joints with modified adhesives demonstrate higher lap shear strength. This investigation was aimed to explore the effect of clay Nano particles (CNP) on the shear strength of glass fiber/epoxy composite single lap joints (SLJs). The shear strength of the lap joints was studied comparatively at different weight ratios of CNP in the adhesive layer. After mechanical tests, Failure loads and effect of additive ratio were presented for each mass rated adhesive with some conclusions.

II. Materials And Methods

Commercial glass fiber composite plates was purchased from local market in Gaziantep, Turkey. Epoxy (MOMENTIVE-MGS L160) and conjugated hardener (MOMENTIVE-MGS H260S) were provided from DOST Chemical Industrial Raw Materials Industry, Turkey. Montmorillonite Nano clay with 35-45 wt. % dimethyl dialkyl (C14-C18) amine was obtained from Grafen Chemical Industries, Turkey. The density of Nano clay is 200-500 kg/m^3 and the Nano particles are in size of 1-10 nm.

Single lap joint configuration was adopted to produce adhesive joints and lap shear test was utilized in order to study the tensile properties of epoxy/Nano composite structures.

2.1 Preparation of Lap Joint Specimens

The composite plates were cut to substrates by guillotine machine in dimensions of 100 × 25 mm according to ASTM D5868-01 standard. The dimensions of SLJs studied are shown in Figure 1. The adhesive layer was based on epoxy and hardener with a weight ratio of 100:28 according to manufacturing specifications. Then, in case of Nano composite adhesive, CNP fillers were added into epoxy by 1, 2, 3, and 5 wt. % (Table 1). The epoxy/clay mixture was stirred for at least 10 min without hardener in order to reach good distribution of CNP. The mixing time increased with the increase of clay amount to avoid particle aggregation. After that the hardener was added and stirring process was continued for extra 2 min in order to prevent temperature rising of the mixture. Before bonding process, substrate surfaces were cleaned with acetone, abraded with a fine abrasive paper (water proof silicon carbide ‘D’166 grade P120D) and then cleaned with acetone again. The measured thickness of composite substrates after abrasion process was 2 mm. An aluminum mold was used to keep substrates in position and to achieve perfect alignment between adherends (Figure 2 (a)). The adhesive was applied on the substrate surfaces within the specified bonding area. Some test samples can be seen in Figure 2 (b).

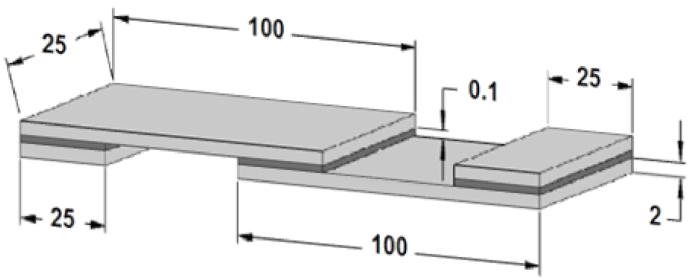

Figure 1. Single Lap Joint Configuration with Dimensions in (mm)

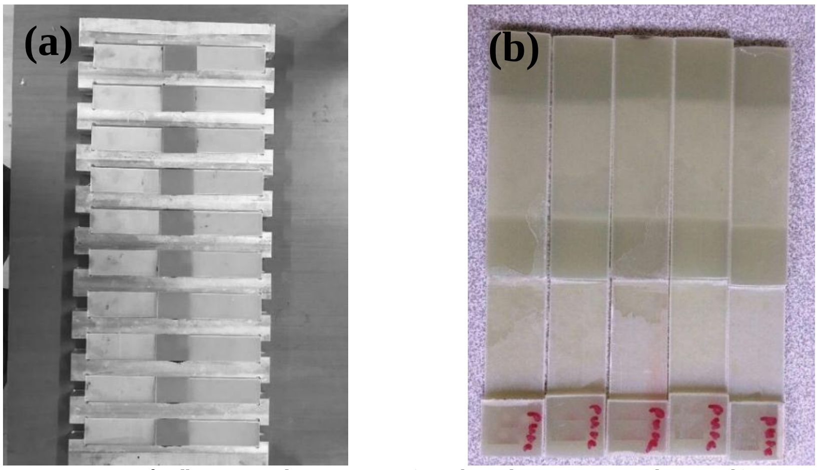

Figure 2. Preparation of Adhesive Single Lap Joints; a) Sticking the Composite Substrates, b) Test Samples

2.2 Lap Shear Test

All the tensile experiments were carried out using a universal computer-controlled tensile device type Shimadzu AG-I (Figure 3 (a)) at room temperature and standard humidity. The cross head speed was 1mm/min according to ASTM D5868-01 standard. The boundary condition and loading are as shown in Figure 3 (b). One end of the lap specimen was pulled away from the other end which was fixed. The overlap length and to the thickness of the adhesive layer of all specimens was fixed. The maximum shear load that the specimens can carry was measured. The SLJ specimens were fractured by subjecting them to shear loads in order to inspect the influence of the CNP reinforcement. Four specimens for each epoxy mixture designed were prepared and tested and the average values were evaluated and adopted. The maximum shear stress (τ_{max}) acting on the adhesively joints is calculated as follows:

$$\tau_{max} = \frac{F_{max}}{bl} \quad (1)$$

Where F_{max} is the maximum load recorded during the lap shear tests, b and l are the width and the length of the overlap area of the joint, respectively.

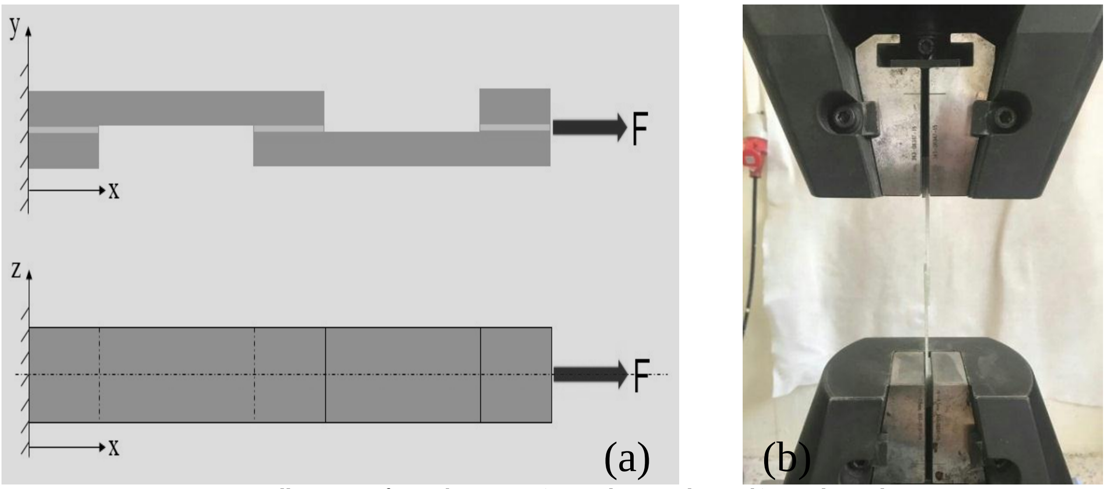

Figure 3. Illustration of Lap Shear Test; a) Boundary Conditions, b) Tensile Loading

III. Results and Discussion

Single lap adhesive bonded joints were prepared with epoxy/CNP composite adhesives and tested under lap shear loading in tension. The joints based on different weight ratios of CNP were tested and compared with control samples of neat epoxy. Two parameters fundamentally dominate the adhesive strength: (a) mechanical characteristics of the epoxy resin and CNP and (b) adhesion properties and viscoelastic behavior of epoxy resin. In previous researches on adhesively joints of epoxy (Saeed and Zhan, 2007), it was noticed that the mechanical properties of the joints increased with the increase of Nano particle content, but, at the same time, their viscoelastic behavior has been changed from liquid-like to solid-like. Thus, it was likely that epoxy containing high weight content of CNP will not have good adhesion properties. The results of lap shear tests in tension on the adhesive joints prepared with epoxy containing different amounts of CNP were obtained and presented in load-displacement diagrams (Figure 4) and shear stress–shear strain diagrams (Figure 5). The shear stress was calculated from tensile load applied on adhesive joints divided by overlap bonding area, and the shear strain was obtained from joint displacement divided by overlap length [28-32]. The maximum shear strength of SLJs (Figure 6) is estimated using the average maximum shear stress shown in Figure 5. The variations of standard deviation for the results shown in Figure 6 were 0.276 to 0.661 MPa for composite epoxy.

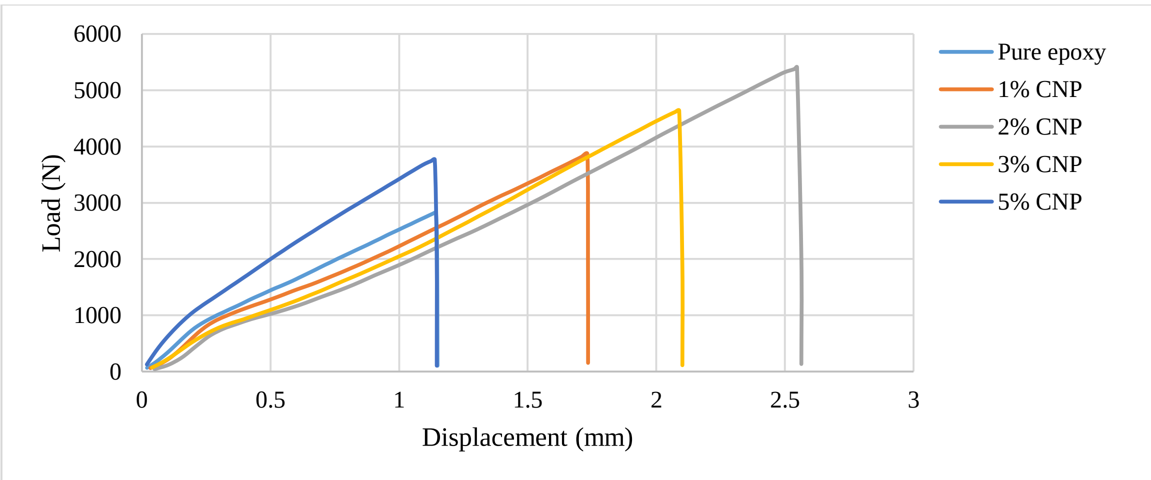

Figure 4. Load-Displacement Curves for SLJs Reinforced with Different Amount of CNP and Subjected to Tensile Loading

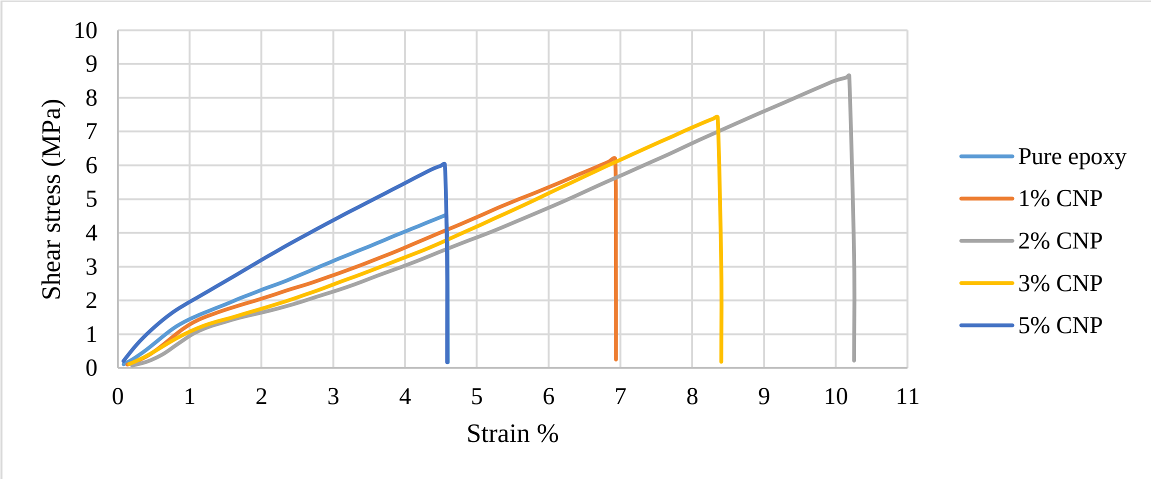

Figure 5. Shear Stress-Shear Strain Curves for SLJs Reinforced with Different Amount of CNP and Subjected to Tensile Loading

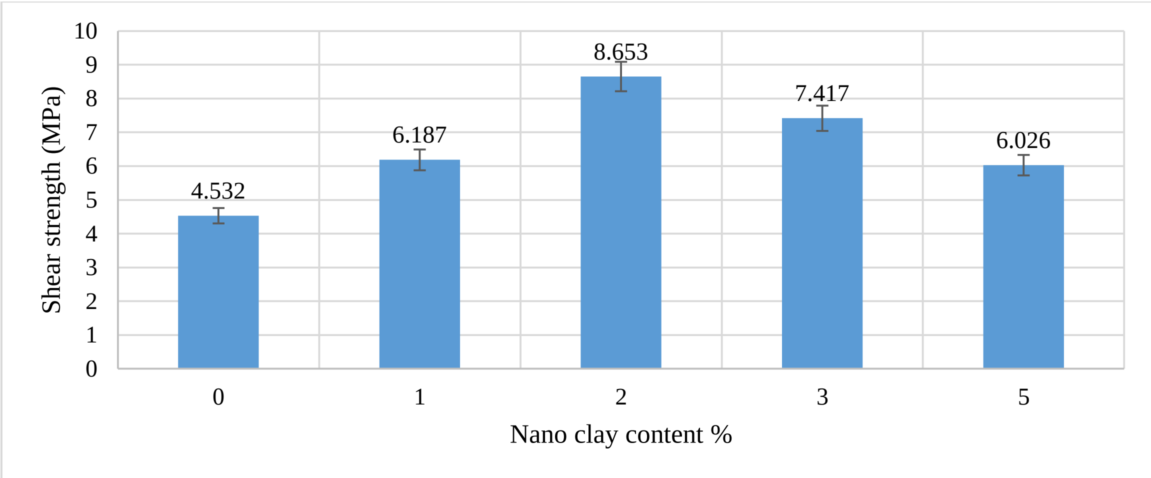

Figure 6. Maximum Shear Strength of SLJs versus CNP Weight Content

The influence of CNP incorporation into the adhesive layer on the shear strength of SLJs was studied comparatively by the results of adhesive joints reinforced and unreinforced with CNP. It can be seen that the shear strength of adhesive joints increased with the addition of CNP, but above 2 wt. % of CNP, the shear strength decreased. The shear strength values for pure epoxy and epoxy reinforced with 1, 2, 3, and 5 wt. % of CNP were 6.187, 8.653, 7.417, and 6.026 MPa, respectively. The results obtained from the lap shear test can be given in Table 1.

TABLE 1. Lap Shear Test Results of SLJs with Various Amount of CNP

Sample code	**Epoxy+Hardener (%)**	**Clay (wt. %)**	**Max. Force (kN)**	**Max. Shear stress (MPa)**	**Max. Shear strain (%)**
T 0	100	0	2832.472	4.532	4.573
T 1	99	1	3867.078	6.187	6.927
T 2	98	2	5408.394	8.653	10.187
T 3	97	3	4635.823	7.417	8.355
T 5	95	5	3766.573	6.026	4.554

The shear strength increased by about 36% and 91% with the addition of 1 and 2 wt. % CNP in the epoxy resin. When CNP content exceeded 2 wt. %, the shear strength of Nano-composite adhesive joints decreased with the increase of Nano clay amount. This can be attributed to the aggregation of particles which causes stress concentration and crack growth in the adhesive layer and lead to sudden failure under lower stresses. However, the results stated that the adhesive joints of Nano composite, for all epoxy/Nano clay designs considered in this study, have shear strength higher than that of control samples. The increase in shear strength was about 36, 91, 63, and 33% for 1, 2, 3, and 5 wt. % of CNP. Much change was noticed in shear strain with the addition of CNP content. The shear strain increased as the CNP increased up to 2 wt. % which resulted in highest shear strain. After that a reduction trend was observed in the shear strain with more addition of CNP than 2 wt. % and the strain value at 5 wt. % was very close to that of neat epoxy. However, for high content ratios, CNP prevent the formation of a homogeneous network in the adhesive layer.

IV. Conclusion

The effect of incorporating clay Nano particles into Epoxy MGS-L160 on the shear strength of SLJs of glass fiber composite samples was investigated. Different weight ratios of Nano particles, namely 1, 2, 3 and 5 wt. % of CNP, were added to the epoxy adhesive in SLJs and the tensile properties were studied. Generally, the experimental results obtained from the axial tensile test and the force-displacement curves for each sample revealed that adding a small weight content of CNP to epoxy adhesive increases the maximum load-carrying capacity by about 91%. The shear strength increased by adding CNP up to 2 wt. % and more than this ratio resulted in decreasing the shear strength. The highest value of shear strength was found for the adhesive joint of epoxy filled with 2% CNP. Strain at break was increased as CNP content increased in the epoxy adhesive up to 2 wt. % and then decreased with more addition of CNP.

REFERENCES

[1] Burkholder, G. L., Kwon, Y. W., & Pollak, R. D. (2011). Effect of carbon nanotube reinforcement on fracture strength of composite adhesive joints. Journal of materials science, 46(10), 3370-3377.
[2] Iqbal, H. M. S., Bhowmik, S., & Benedictus, R. (2014). Process optimization of solvent based polybenzimidazole adhesive for aerospace applications. International Journal of Adhesion and Adhesives, 48, 188-193.
[3] zirconia nanoparticles. Composite Interfaces, 17(9), 873-892.
[4] Dorigato, A., Pegoretti, A., Bondioli, F., & Messori, M. (2010). Improving epoxy adhesives with Armstrong, K. B. (1997). Long-term durability in water of aluminium alloy adhesive joints bonded with epoxy adhesives. International Journal of Adhesion and Adhesives, 17(2), 89-105.
[5] Armstrong, K. B., Bevan, L. G., & Cole, W. F. (2005). Care and repair of advanced composites. Society of Automotive Engineers.
[6] Meguid, S. A., & Sun, Y. (2004). On the tensile and shear strength of nano-reinforced composite interfaces. Materials & design, 25(4), 289-296.
[7] Saeed, M. B., & Zhan, M. S. (2007). Adhesive strength of nano-size particles filled thermoplastic polyimides. Part-I: Multi-walled carbon nano-tubes (MWNT)–polyimide composite films. International Journal of Adhesion and Adhesives, 27(4), 306-318.
[8] Xu, S., Dillard, D. A., & Dillard, J. G. (2003). Environmental aging effects on the durability of electrically conductive adhesive joints. International Journal of Adhesion and Adhesives, 23(3), 235-250.
[9] Vinson, J. R., & Sierakowski, R. L. (1987). The Behavior of Structures Composed of Composite Materials. Martinus Nijhoff, Dordrecht: Kluwer–Academic.
[10] Fayad, M.A., Tsolakis, A. and Martos, F.J., 2020. Influence of alternative fuels on combustion and characteristics of particulate matter morphology in a compression ignition diesel engine. Renewable Energy, 149, pp.962-969.
[11] Fayad, M.A., 2012. Analytical solution for predicting heat pipe performance. AL-TAQANI, 25(1), pp.E-82.
[12] Hamidi, S.T. and Fayad, M.A., 2011. Theoretical study for aerodynamic performance of horizontal axis wind turbine. AL-TAQANI, 24(8), pp.E113-E124.
[13] Al Salihi, H.A., Fayad, M.A., Slepchenkov, M. and Shunaev, V., 2020, April. Nanoscale oscillator on the base of single-walled carbon nanotube with internal fullerenes C36 and C80. In Saratov Fall Meeting 2019: Laser Physics, Photonic Technologies, and Molecular Modeling (Vol. 11458, p. 1145811). International Society for Optics and Photonics.
[14] Ferreira, J. A. M., Reis, P. N., Costa, J. D. M., & Richardson, M. O. W. (2002). Fatigue behaviour of composite adhesive lap joints. Composites Science and Technology, 62(10), 1373-1379.
[15] Gilbert, E. N., Hayes, B. S., & Seferis, J. C. (2003). Nano-alumina modified epoxy based film adhesives. Polymer Engineering & Science, 43(5), 1096-1104.
[16] Gojny, F. H., Wichmann, M. H., Fiedler, B., & Schulte, K. (2005). Influence of different carbon nanotubes on the mechanical properties of epoxy matrix composites–a comparative study. Composites Science and Technology, 65(15), 2300-2313.
[17] Guadagno, L., Sarno, M., Vietri, U., Raimondo, M., Cirillo, C., & Ciambelli, P. (2015). Graphene-based structural adhesive to enhance adhesion performance. RSC Advances, 5(35), 27874-27886.
[18] Isik, I., Yilmazer, U., & Bayram, G. (2003). Impact modified epoxy/montmorillonite nanocomposites: synthesis and characterization. Polymer, 44(20), 6371-6377.
[19] Kim, J. K., Hu, C., Woo, R. S., & Sham, M. L. (2005). Moisture barrier characteristics of organoclay–epoxy nanocomposites. Composites Science and Technology, 65(5), 805-813.
[20] Li, X., Gao, H., Scrivens, W. A., Fei, D., Thakur, V., Sutton, M. A., Myrick, M. L. (2005). Structural and mechanical characterization of nanoclay-reinforced agarose nanocomposites. Nanotechnology, 16(10), 2020.
[21] Lin, J. C., Chang, L. C., Nien, M. H., & Ho, H. L. (2006). Mechanical behavior of various nanoparticle filled composites at low-velocity impact. Composite Structures, 74(1), 30-36.
[22] Liu, W., Hoa, S. V., & Pugh, M. (2005). Fracture toughness and water uptake of high-performance epoxy/nanoclay nanocomposites. Composites Science and Technology, 65(15), 2364-2373.
[23] Fayad, M.A., 2020. Effect of renewable fuel and injection strategies on combustion characteristics and gaseous emissions in diesel engines. Energy Sources, Part A: Recovery, Utilization, and Environmental Effects, 42(4), pp.460-470.
[24] Fayad, M.A., Tsolakis, A., Fernández-Rodríguez, D., Herreros, J.M., Martos, F.J. and Lapuerta, M., 2017. Manipulating modern diesel engine particulate emission characteristics through butanol fuel blending and fuel injection strategies for efficient diesel oxidation catalysts. Applied Energy, 190, pp.490-500.
[25] Fayad, M.A., Herreros, J.M., Martos, F.J. and Tsolakis, A., 2015. Role of alternative fuels on particulate matter (PM) characteristics and influence of the diesel oxidation catalyst. Environmental science & technology, 49(19), pp.11967-11973.
[26] Miyano, Y., Nakada, M., Ichimura, J., & Hayakawa, E. (2008). Accelerated testing for long-term strength of innovative CFRP laminates for marine use. Composites Part B: Engineering, 39(1), 5-12.
[27] Patel, S., Bandyopadhyay, A., Ganguly, A., & Bhowmick, A. K. (2006). Synthesis and properties of nanocomposite adhesives. Journal of adhesion science and technology, 20(4), 371-385.
[28] Fayad, M.A., Fernandez-Rodriguez, D., Herreros, J.M., Lapuerta, M. and Tsolakis, A., 2018. Interactions between aftertreatment systems architecture and combustion of oxygenated fuels for improved low temperature catalysts activity. Fuel, 229, pp.189-197.
[29] Bogarra-Macias, M., Doustdar, O., Fayad, M.A., Wyszyński, M.L., TSOLAkiS, A., Ding, P., Pacek, A., MARTiN, P., Overend, R. and O'Leary, S., 2016. Performance of a drop-in biofuel emulsion on a single-cylinder research diesel engine. Combustion Engines, 55.
[30] Fayath, M.A. and Hamidi, S.T., 2011. Prediction of thermal characteristics for solar water heater. Anbar Journal of Engineering Sciences, 4(2), pp.18-32.
[31] Prolongo, S. G., Gude, M. R., & Ureña, A. (2010). Nanoreinforced adhesives. In Nanofibers. InTech.
[32] Ragosta, G., Abbate, M., Musto, P., Scarinzi, G., & Mascia, L. (2005). Epoxy-silica particulate nanocomposites: chemical interactions, reinforcement and fracture toughness. Polymer, 46(23), 10506-10516.
[33] Sarathi, R., Kumar, P. R., & Sahu, R. K. (2007). Analysis of surface degradation of epoxy nanocomposite due to tracking under AC and DC voltages. Polymer degradation and stability, 92(4), 560-568.
[34] Silva-Neto, A., Cruz, D. T. L. D., & Ávila, A. F. (2013). Nano-modified adhesive by graphene: the single lap-joint case. Materials Research, 16(3), 592-596.
[35] Varghese, S., Gatos, K. G., Apostolov, A. A., & Karger-Kocsis, J. (2004). Morphology and mechanical properties of layered silicate reinforced natural and polyurethane rubber blends produced by latex compounding. Journal of applied polymer science, 92(1), 543-551.
[36] Wernik, J. M., & Meguid, S. A. (2014). Multiscale micromechanical modeling of the constitutive response of carbon nanotube-reinforced structural adhesives. International Journal of Solids and Structures, 51(14), 2575-2589.
[37] Xi, X., Yu, C., & Lin, W. (2009). Investigation of nanographite/polyurethane electroconductive adhesives: preparation and characterization. Journal of Adhesion Science and Technology, 23(15), 1939-1951.
[38] Yao, X. F., Yeh, H. Y., & Zhao, H. P. (2005). Dynamic response and fracture characterization of polymer-clay nanocomposites with mode-I crack. Journal of composite materials, 39(16), 1487-1496.

[39] Yu, S., Tong, M. N., & Critchlow, G. (2009). Wedge test of carbon-nanotube-reinforced epoxy adhesive joints. Journal of Applied Polymer Science, 111(6), 2957-2962.
[40] Zhai, L. L., Ling, G. P., & Wang, Y. W. (2008). Effect of nano-Al 2 O 3 on adhesion strength of epoxy adhesive and steel. International Journal of Adhesion and Adhesives, 28(1), 23-28.
[41] Fayad, M.A., 2019. Effect of fuel injection strategy on combustion performance and NO x/smoke trade-off under a range of operating conditions for a heavy-duty DI diesel engine. SN Applied Sciences, 1(9), p.1088..
[42] Fayad, M.A. and AL-Ogaidi, B.R., 2019. Investigation the Morphological Characteristics of the Particulate Matter Emissions from the Oxygenated Fuels Combustion in Diesel Engines. Engineering and Technology Journal, 37(10 A), pp.384-390.
[43] Fayad, M.A., 2017. Particulate Matter (PM) characteristics from compression ignition diesel engines operated by renewable fuels (Doctoral dissertation, University of Birmingham).
[44] Farge, T.Z., Shibib, I.I., Fayad, M.A. and Alwan, M.A., 2011. Per Formance Improvement Of A Turbo Shaft Engine Using Water Injection. Iraqi journal of mechanical and material engineering, 11(3), pp.417-436.
[45] Salman, A.Z., Fayad, M.A. and Salam, A.Q., Theoretical Investigation of Buoyancy-driven Cavity Flow by Using Finite Element.
[46] Zhai, L., Ling, G., Li, J., & Wang, Y. (2006). The effect of nanoparticles on the adhesion of epoxy adhesive. Materials Letters, 60(25), 3031-3033.
[47] Zhang, J., Jiang, D. D., & Wilkie, C. A. (2006). Fire properties of styrenic polymer–clay nanocomposites based on an oligomerically-modified clay. Polymer degradation and stability, 91(2), 358-366.