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Increasing the Polarity of the Aluminum Surface and TPENR Matrix with the Addition 3-GPS and PE-g-MAH as a Coupling Agent to Improve the Lap Shear Strength of Al-TPENR Laminated Composite

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ABSTRACT—*In this study, we analyzed the essence of surface treatment of the aluminium surface and a coupling agent to better adhesion between aluminium with thermoplastic epoxidized natural rubber (TPENR). Thermoplastic epoxidized natural rubber (TPENR) matrix was prepared by melt blending of linear low density polyethylene (LLDPE), epoxidized natural rubber (ENR), liquid epoxidized natural rubber (LENR) as a compatibilizer, and polyethylene grafted Maleic anhydride (PE-g-MAH) as a coupling agent. The PE-g-MAH concentration used was varied from 0% - 30%. In addition, the aluminium surface was pre-treated with 3-glycidoxy propyl trimethoxy silane (3-GPS) to enhance the mechanical properties and chemical interaction of laminated composite. It was found that the shear strength of the single lap joint Al-TPENR laminated composite showing an increasing trend as a function of PE-g-MAH contents for the 3-GPS surface treated aluminium. Moreover, the FTIR-ATR, XPS and SEM analysis revealed that the strength improvement was linked up with the chemical state of the compound involved.*

Keywords—Shear strength, single lap joint, and laminated composite of Al-TPENR.

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1. INTRODUCTION

Recently, a lot of attention has been focused upon the development of polymer matrix composite with metals (sandwich/laminate composite). These composites are used in a wide range of applications such as aerospace, automotive, implants, electronic, packaging, defense, nuclear, medical, etc[1,2]. As compared to homogenous metal sheets, the metal-polymer laminated composite have many unique properties in terms of low density and weight, high bending strength as well as good sound, vibration and damping characteristics [1-5].

The preparation of suitable metal-polymer laminates requires many practical considerations. It has been established that one of the most important considerations is the pretreatment process of metal surface either by mechanical, chemical, and electrochemical nor the coupling agents. The coupling agent is well-known to enhance the molecular bonding between polymer matrix and inorganic particulates. It strengthens the interfacial adhesion when one end of the molecule is tethered to the reinforcement surface and the functionality at the other end reacts with the polymer phase [6-10,11-13]. Typically, the coupling agents are classified into organic, inorganic, and organic-inorganic groups. The organic agents include isocyanates, anhydrides, amides, imides, acrylates, chlorotriazines, epoxides, organic acids, monomers, polymers, and copolymers. On the other hand, the inorganic agents such as silicates can be found in the study of nanocompsite (Polymer-layered silicate nanocomposites). The other types include the organic-inorganic agents, i.e. silane and titanate [14].

It is a tedious and time-consuming process to select the appropriate coupling agents experimentally. Silane is recognized as an efficient coupling agent that can be used in the composites and adhesive formulations [15]. Silane coupling agents have a generic chemical structure $R_{(4-n)}{-}Si{-}(R'X)_n$ ($n = 1, 2$) where R is alkoxy, X represents an organo-functionality, and R' is an alkyl bridge connecting the silicon atom and the organofunctionality. The organofunctionality for every silanes is very specific. Most of them were used during the past decade by the researchers are amino, APS [16], mercapto, MRPS [17], glycidoxy, GPS [18,19], vinyl, VTS [20], alkyl, HDS [21], or methacryloxy group, MPS[22].

Usually polar materials with good strength are selected as a polymer matrix. Functional groups like –C=O, –OH, –COOH, $-NH_2$, etc., make a polar polymer with inherent surface strength better than the non-polar polymers [4,9,11]. The non-polar polymers can be surface modified by surface oxidizing [23], chemical etching [24], surface grafting [25], flame treatment [26], electron beam [27], microwave irradiation [28], electrical plasma discharge [29] and glow discharge [30] etc. All of these processes aim to create polar groups at the polymer surface to increase the value of the surface-free energy and adhesive properties of polymeric materials. In recent years, many studies are dedicated to obtain new materials by blending between polyolefins with natural materials that is rich in functional groups like –OH, –CO, –CHO and etc., For example, the use of starch [31], wood [32], rice straw [33] etc., and natural rubber [34]. Blending between polyolefin as a thermoplastic polyolefin (TPO) with natural rubber is known as thermoplastic elastomer (TPE) but it is commonly called as a thermoplastic natural rubber (TPENR).

Apart from modifying the polymer surface, the adhesion between polymeric material with the primary metal structures also requires some attention. It is well-known that the polyethylene exhibits insufficient adhesive bond strength due to low surface energy. This property drawback has been solved by chemical, thermal, mechanical and electrical treatments. However, the dry treatment method is more suitable for industrial applications. Previous works have shown the effect of the maleic anhydride coupling agent [2,9,11] by imparting polarity to a polyolefin surface that enables an improved adhesive bonding with aluminium and pretreated aluminium surface [8,10].

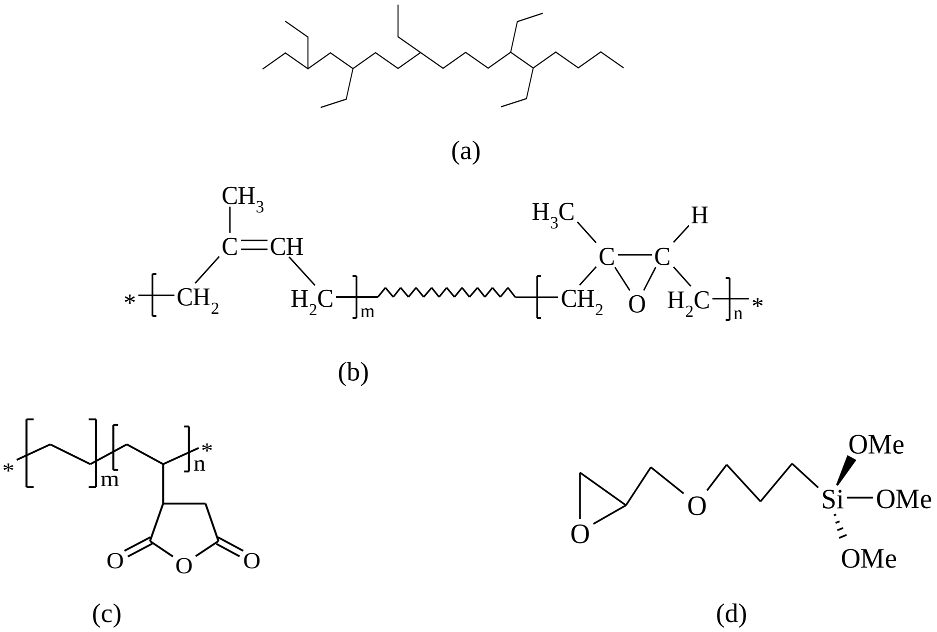
Chen et al. [1] prepared an aluminum-polypropylene (Al-PP) laminated composite by using PP-g-MAH as a coupling agent and 3-APS was used to treat the surface of the aluminum. Da silva et al. [35] fabricated steel-epoxy composite laminates and steel surfaces coated with an organic phosphate, while Pereira et al. [36] fabricated Al-epoxy composite laminates and then aluminum surface treated with sodium dichromate-sulfuric acid etch (CSA). Putman and Vaida [2] studied mechanisms of interfacial adhesion in metal–polymer composites. They produced a steel-polymer laminate composite from their studies. Polymer laminates are hard TPU Pearlthane D11T75 merquinsa, soft TPU Pearlthane D11T85E merquinsa, nylene Custom Resins 401D, PP, and PPMA as a coupling agent. Steel surface was coated with an adhesive nano fullerene epoxy resin CNT epoxy. Sokolova et al. [37] produced a composite laminate consisting of high-quality austenitic stainless steel (316L) sheets with anominal thickness of 0.5 mm. The core consisted of a polyolefin foil being a mixture of polypropylene (PP) and polyethylene (PE) polymers, talc $[Mg_3Si_4O_{10}(OH)_2]$, rutile (TiO_2) and barite ($BaSO_4$). Recently, Zhang et al. [38] produced a composite laminate of aluminum alloy-epoxy-soft steel. The main ingredient was the matrix diphenol epichlorohydrin-4,4-isopropylidene with acrylic butadiene copolymer (CTBN) as a reinforcing agent.

In the present work, we investigated the potential use of polyethylene grafted maleic anhydride (PE-g-MAH) coupling agent in the thermoplastic natural rubber (TPENR) LLDPE/ENR/LENR matrix for Al-TPENR laminated composite. In the addition, the 3-GPS used as pretreatment to the surface of ground aluminium sheet in order to improve the shear strength of single lap joint Al-TPENR laminated composite.

2. EXPERIMENTAL

2.1 Material

Epoxidized natural rubber (ENR) was obtained from the Rubber Research Institute of Malaysia (RRIM). Liquid of epoxidized natural rubber (LENR) was prepared in our laboratory by photochemical oxidation of epoxidized natural rubber (ENR). Natural rubber (1 kg) was cut approximately into 1 cm × 1 cm × 1cm cube. After that, it was mixed in toluene solvent with 0.2 g methylene blue, 0.26 g rose bengal and 10 mL methanol. Liquid natural rubber was formed when exposed under UV in a photoreactor after 12 days [39]. Linear low density polyethylene (LLDPE) from Exxon Mobil Chemical Manufacturing Singapore was used. The metal used is a pure aluminium sheet with specifications; thickness 1.5 mm, serial number AA1100 (Al = 99.00%; Si & Fe = 0.05%, Mn = 0.05%; Zn = 0.10%; Cu = 0.05-0.2% and other content = 0.05%). Other materials used are polyethylene-grafted-maleic anhydride, PE-g-MAH (melting point = 105 °C; d = 0.925 g/mL) and silane coupling agent of 3-glycidoxy-propyl-trimethoxy-silane (3-GPS), $[(H_2COCH)\text{-}CH_2O(CH_2)_3Si(OCH_3)_3]$, both are manufactured in Aldrich Chemistry USA. The chemical structure of PE-g-MAH and 3-GPS are shown in **Figure 1**.



(a)

(b)

(c)

(d)

Figure 1: Chemical structure of; (a) LLDPE, (b) epoxidized natural rubber (ENR), (c) PE-g-MAH and (d) 3-GPS

2.2 Preparation of Al-TPENR laminated composite

Thermoplastic epoxidized natural rubber (TPENR); LLDPE, ENR, LENR (80 : 15 : 5) and PE-g-MAH were blended by using an internal mixer, ThermoHaake, at 140 °C, rotor speed 50 r.min^{-1} and mixing time of 12 minutes. Various concentrations of PE-g-MAH (0, 5, 10, 15, 20, 25, and 30%) were incorporated into the compounds. The matrix is made of thin layers using a compression molding at a temperature of 160 °C, pressure 0.5 MPa for 10 min. Aluminium sheets were cut to single lap shear tensile test, following to the standard of ASTM D 1002-72 for the sandwich structure. The aluminium sheets were boiled in distilled water for 30 min, cleaned with acetone at room temperature, and then ground with sandpaper of P600. After that, the aluminium sheets were cleaned again with acetone at room temperature and then dried in the ambient air. Several sheets of aluminium were treated by dipping into a solution of 3-GPS 1% (in ethanol), for 5 minutes, was stirred continuously for another 30 minutes. The sample matrix was then placed between two aluminium sheets and compressed to 10 minutes under the pressure of 0.5MPa, at 160 °C temperatures. The thicknesses of the matrix (TPENR) between two aluminium sheets are controlled to be about 0.05 mm by the applied pressure and moulding.

2.3 Characterization of Al-TPENR laminated composite

Tensile properties were measured using 10-tons Testometric M500-50CT tester with a crosshead speed of 1 mm/min. Every value of lap shear strength was averaged of 5 specimens include standard error. Fracture surface morphology of single lap shear tensile sample was carried out by using LEO 1450VP scanning electron microscope (SEM) with an acceleration voltage of 20kV. Various elements, functional groups and bonds formed on the top surface of the aluminium were investigated using X-ray photoelectron spectroscopy (XPS) AXIS Ultra DLD with monochromatic Al Kα radiation source (1486.6 eV), operating at a power of 150 W, was used to generate the emission of photoelectrons from the surface of the samples. Fourier transforms infrared (FTIR) spectroscopy spectrum 400 with attenuated total reflectance (ATR) method analysis to determinate functional group on the surface. For each spectrum, ATR ranges 4000 cm^{-1} to 650cm^{-1}, 10 scans at a spectral 4 cm^{-1} resolution were recorded.

3. RESULT AND DISCUSSION

3.1 FTIR spectroscopy analysis

Figure 2 shows FTIR spectra of LLDPE, PE-g-MAH, ENR 50, TPENR (LLDPE/ ENR/LENR) and TPENR plus PE-g-MAH (LLDPE/ENR/LENR/PE-g-MAH). The functional groups that are related to the chemical state of Al-TPENR laminated composite interfaces such as –C=O can be characterized in the frequency 1709 cm^{-1} (for PE-g-MAH), 1645 cm^{-1} (for matrix system LLDPE/ENR/LENR) and –OH in the frequency 3394 cm^{-1} respectively. Both of the matrixes TPENR and TPENR/PE-g-MAH also have a peak C–H, $-CH_2$ and C–O–C (epoxy or anhydride ring) at wavenumbers 2847 and 2914, 1462, 880 cm^{-1} respectively. The appearance of functional groups –C=O and –OH on the matrix surface were believed to have formed during mixing as a result of material oxidation. These functional groups are important in the physical and chemical interactions in the bonding interface of Al-TPENR laminated composite [1,2].

Chen et al. [9] reported that carboxylate complexes (Al–O–C=O) formed through interfacial interaction of MAH with metal substrates by Fourier transform infrared spectroscopy (FTIR) techniques and also proposed that carboxylate complexes formed by interfacial interaction between MAH grafted in PP and surfactant on the aluminium surface as a substrate. Yang et al. [19]reported that kinetics and reaction mechanism of γ-GPS on low carbon steel surfaces by FTIR-ATR, AFM, NSS and theoretical calculation method. They reported that the reaction of γ-GPS on low carbon steel surfaces followed the conventional reaction mechanism, which can be described as a reaction (I) (Me (Metal)–OH + HO–Si → Me–O–Si + H_2O) and reaction (II) (Si–OH + Si–OH → Si–O–Si (siloxane) + H_2O).

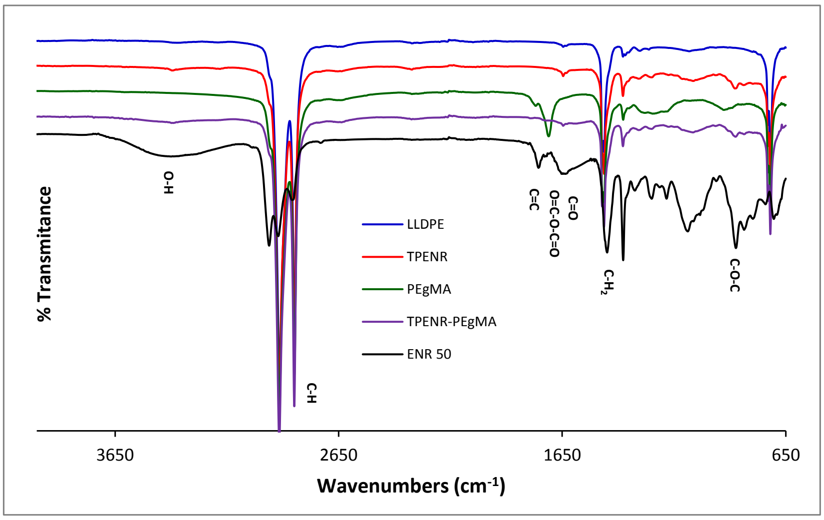
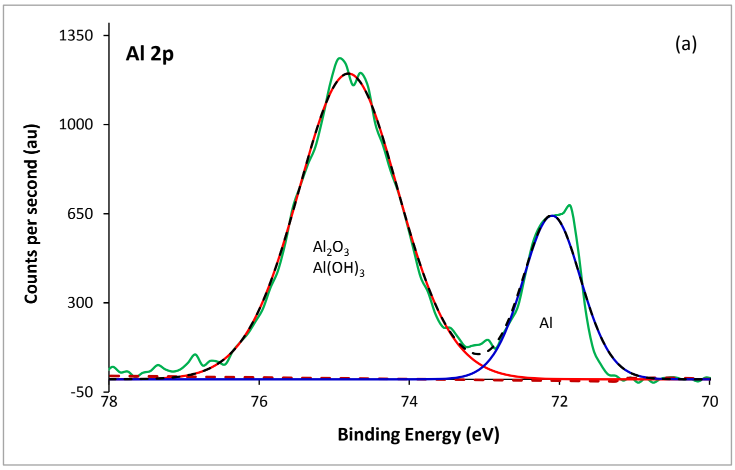
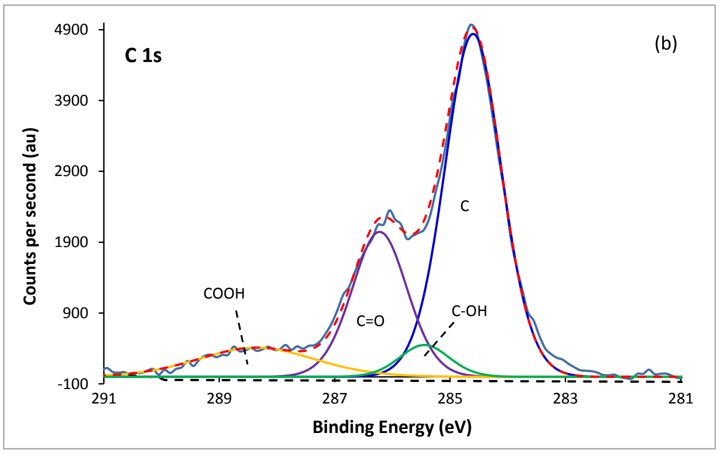


Figure 2: FTIR spectra for LLDPE, PEgMAH, TPENR, TPENR/PE-g-MAH and ENR 50

3.2 XPS analysis

An XPS spectrometer of the aluminium surface after the pretreatment procedure is shown in the **Figure3** (a, b, c and d). Al 2p spectra after pretreatment showed the formation of Al (72.2 eV), Al_2O_3 (74.4 eV) and 74.8 eV for Al-OH, C 1s showed formation of C-C and CH (284.7 eV), COH (285.6 eV), COC and C=O (286.4 eV) and 288.5 eV for COOH, O 1s showed formation O= or M=O (532.2 eV), −O− or M-OH (533.7 eV) and 534.3 eV for H_2O, and Si 2p also showed formation Si element (99.5 eV), Si (silane) (101.5 eV) and 103.7 for SiO_2 at the surface of aluminum. The formation of Al-OH or COOH functional groups on the surface of the aluminium, resulted in physical interaction, such as the van der Waals and hydrogen bonding interaction between TPENR or PE-g-MAH and surfactants (3-GPS) of the pretreated aluminium sheets at the interface. This has contributed to improve the interfacial adhesive strength and the chemical interactions at the interface [2,6,8,9,12].


Al 2p
(a)
Counts per second (au)
1350
1000
650
300
-50
Al_2O_3
$Al(OH)_3$
Al
78
76
74
72
70
Binding Energy (eV)


C 1s
(b)
Counts per second (au)
4900
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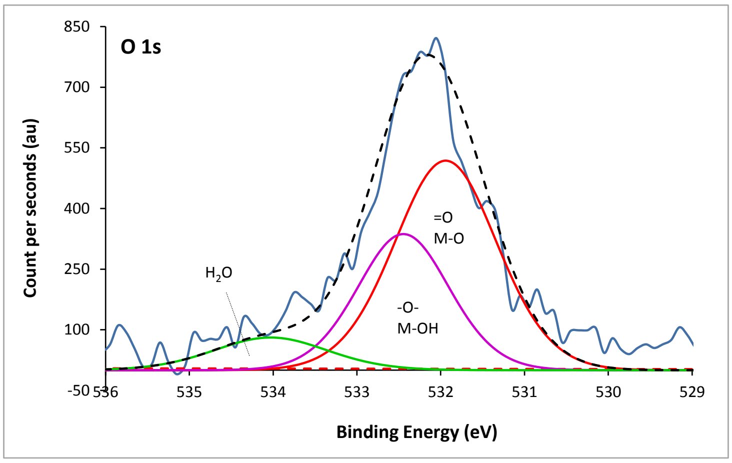
Binding Energy (eV)


O 1s
Count per seconds (au)
850
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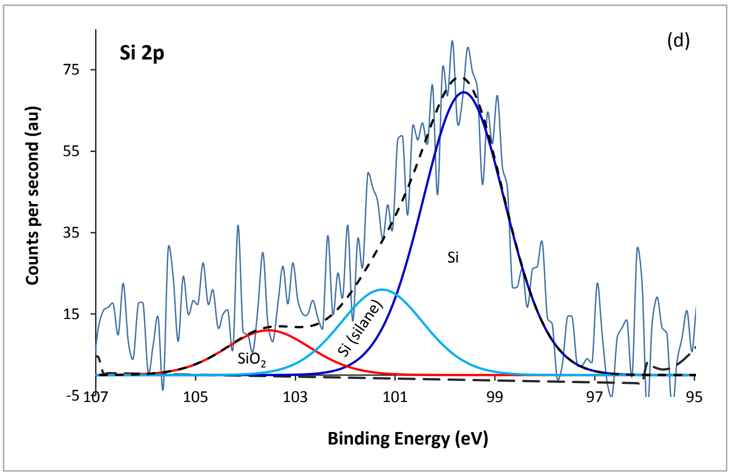
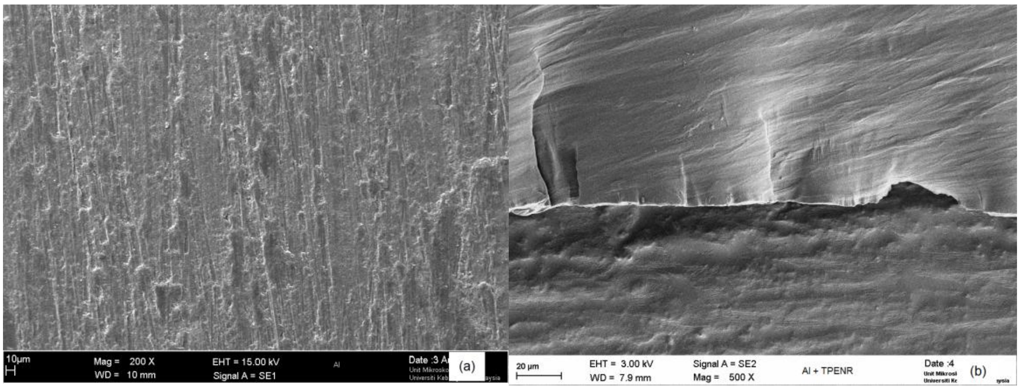


Figure 3: XPS spectra of on the aluminium surface with 3-GPS, separated polymer surface Al-TPENR laminated composite; (a) Al 2p, (b) C 1s, (c) O 1s, and (d) Si 2p

Chen et al. [1] reported that the effect of the amount of maleic anhydride-grafted polypropylene (PP-g-MAH) in polypropylene (PP) and aluminium surface pretreated by 3-aminopropyltrimethoxysilane (3-APS) on lap shear strength of adhesive-bonded Al-PP laminated composite. For pure PP, the lap shear strengths were found to be weaker than composites. SEM analysis has shown that a fracture with almost no plastic deformation on the aluminium surface was observed. Modification of PP by the addition of 5 – 30 wt% amount of PP-g-MAH increased the lap shear strength significantly. It was due to the chemical interactions between –OH, Al^{3+} or amino group $–NH_2$ at the surface of the aluminium sheets and the polar functional anhydride groups and carboxylic groups –COOH on PP-g-MAH at the interface (polar group investigated by XPS analysis).

3.3 SEM analysis

Figure 4 (a, b, c and d) shows the SEM micrographs of the lap shear fractured aluminium surfaces with and without PE-g-MAH in TPENR matrix and coupling agent, 3-GPS. It can be seen in Figure 4 (a, b) that almost all aluminum surfaces are clearly visible and almost no inherent TPENR and also figure 4 (b) shows the surface of TPENR matrix easily separated from the aluminum metal, it is due to no interfacial physical and chemical interaction. In contrast, when the PE-g-MAH was added and aluminum surfaces protected with 3-GPS, it comes out the fracture surface of aluminum remains as a strong matrix inherent as shown in Figure 4 (c) and adhesion between the aluminum metal matrix can be viewed from the side lap joint in Figure 4 (d). Chen et al [1] reported that pretreatment aluminum surface with the addition of coupling agent and maleic anhydride grafted into the plastic has enhanced the adhesion strength between polymer matrices with metal; hence an improved composite laminate could be obtained [9,12].



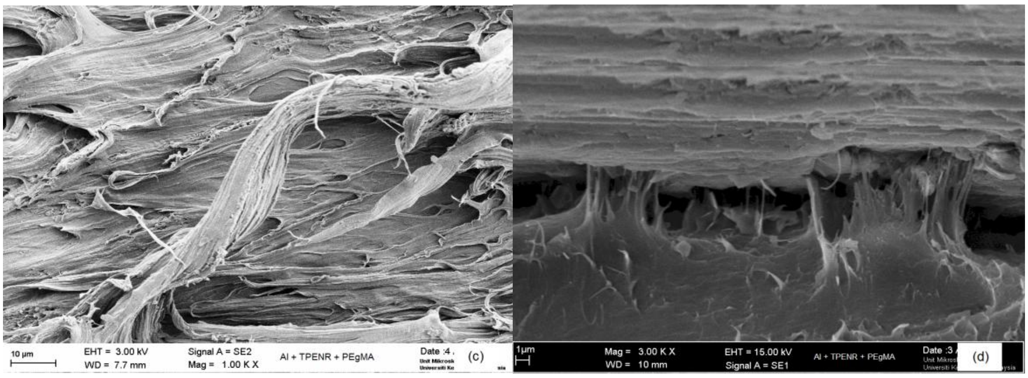


Figure 4:SEM images of lap shear fractured surface; (a) of the adhering aluminum sheet without 3-GPS and PE-g-MAH, (b) adhered aluminium sheet with 3-GPS and PE-g-MAH 1 wt% added in TPENR matrix.

3.4 Lap Shear Strength

Figure 5 shows the shear strength of the single lap joint Al-TPENR laminated composite with TPENR plus PE-g-MAH and the coupling agent, 3-GPS, which was added as a pretreatment on the surface of the aluminium sheet. With the addition of 25% PE-g-MAH into the TPENR, the lap shear strength increased from 0.8 MPa to 8.0 MPa as compared to TPENR system only. This remarkable increment of 943% was further enhanced for the laminate composite treated with 3-GPS 1% coupling agent. The lap shear strength was further increased to 9.8% against the untreated laminate composite. This could be contributed by the physical interactions, such as the van der Waals interaction between TPENR and surfactants of the pretreated aluminium sheets as well as the mechanical interlocking between TPENR and surface roughness of the aluminium sheets at the interface. With the addition of grafting maleic anhydride into polyolefin and coupling agent on the aluminium surface, the increase in the lap shear strength should be ascribed to the contribution of chemical interactions at the interface [1,5,7,11,12].

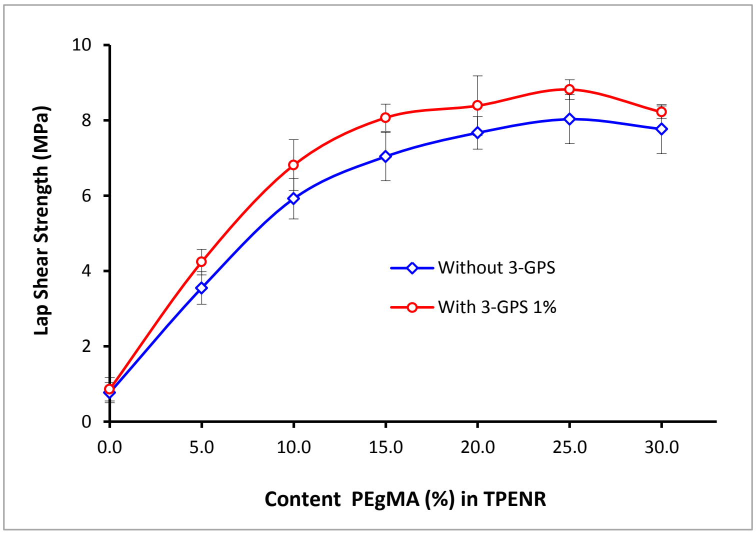


Figure 5: The effect of the PE-g-MAH content (%) on the shear strength of the single lap joint of Al-TPENR laminated composite: (–o–) with pretreatment 3-GPS 1% solution, (–◊–) without pretreatment by 3-GPS solution.

The shear strength of the single lap joint of laminate composite was performed to analyze the effect of surface roughness, polymer type and metal surface treatment to the bond strength between metal and polymer (interface adhesion mechanism).It has been widely postulated that the creation of covalent bonds at the interface is sufficient for creating viable adhesive strength in adhesion-related applications [1,6,11].

The contribution in the lap shear strength laminated composite is not only due to the chemical interactions and physical strength at the interface, but also the procedure of curing. The physical and mechanical properties of adhesives, adhesive and bonding stiffness base metal interface also contributes the strength of the laminate composite. You et al. [40] proposed that the failure mode in the single lap joint may be changed with the curing procedure or conditions, physical and mechanical properties of the epoxy adhesive, and the steel base interface. By changing the extent of adhesive curing or interface properties, the stress distribution within the joint could be modified. Reis et al. [41] reported that the shear strength of lap joints was significantly influenced by stiffness, and the highest shear stresses were obtained by using high stiffness adherend materials. The superposition length influences the shear strength in different ways depending on the different adherend materials.

Figure 5 shows that after the addition of PE-g-MAH at 25% concentrations, the shear strength decreases. This phenomenon occurs due to capacity (concentration) matrix interaction with the surface. Generally, the interface bonding surface strength increases monotonically with the number of interactions. Bonding strength can be determined by capacity surface bonding and interface mechanism. Song et al. [42] reported that adhesion and frictional performances of the molecule or functional groups textured surfaces strongly correlated to the wetting property of the textured surfaces that is the larger thewater contact angle (WCA), the better the adhesion and friction performances of the textured surfaces. When the concentration of PE-g-MAH has reached a critical concentration, the excess concentration of PE-g-MAH is no longer interacting with the metal surface but interacts with the nonpolar from the other side of the PE-g-MAH. The interaction force between –COOH functional groups of PE-g-MAH with $-CH_3$ in polymer or PE-g-MAH is a weak interaction compared interaction –COOH with –OH on the surface or –COOH group on PP [43]. Ku et al [44] reported that the shear strength of aluminum-polypropylene laminated composite increased after surface oxidation of polypropylene by atmospheric pressure plasma method. Increased PP surface roughness and concentration of hydrophilic functional groups −CO and −OH generated after plasma, indicated the formation of the chemical and physical interactions at the surface of the PP with aluminium.

Author names are to be centered beneath the title and typed in Times New Roman 11-point, non-italic, non-boldface type. Multiple authors may be shown in a two or three-column format, with their affiliations below their respective names. Affiliations are centered below, typed in Times New Roman 10-point non-italic, non-boldface. Include e-mail addresses if possible typed in Times New Roman 10-pointitalic, non-boldface. Follow the author information by two blank lines before main text.

4. CONCLUSIONS

The shear strength of single lap joint Al-TPENR laminated composite increased by 943% when the PE-g-MAH was added at 25% into the TPENR. With the 3-GPS 1% coupling agent on the treated aluminium surface, the lap shear strength was further increased by another 9.8% as compared to the untreated laminate composite. These phenomena could be associated with the physical and chemical interaction between the Al-OH, COOH, CO, COC, COH and AlOSi as confirm by the XPS, FTIR and SEM analysis.

5. ACKNOWLEDGEMENT

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