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Interlaminar Shear Behaviour of UHMWPE (Ultrahigh Molecular Weight Polyethylene) Based Composites with Different Matrixes

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Abstract: Ultrahigh molecular weight polyethylene (Endumax®) based composite was treated to investigate the route to improve interlaminar shear strength (ILSS) and its failure mechanism. The failure mechanism of the composite was observed in this study by some failure tests such 3-point bending on composite, and SEM analysis. To increase ILSS value of this UHMWPE based composite, some routes were done by using different matrixes (HDPE film, LLDPE film, Epoxy resin, Styrene). For overall practical approach, the failure mechanism detected in this study was caused by delamination of UHMWPE fibrils. They were ripped apart during mechanical test that lead to delamination of the composite. Numerous practical approach conducted, but the UHMWPE composite would keep at low shear strength as long as nothing enhance the interaction between the UHMWPE fibrils.
Keywords: Endumax®; UHMWPE; matrix; Interlaminar shear; Failure mechanism
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1. Introduction

Replacement of metals, especially steel and iron has been a popular topic for technical textiles area which replaced with high performance fibres based composite. According to Ajayan and Tour, composite is defined as material formed from two or more distinct materials, have desirable combination of properties that aren't found in the individual component [1]. The composite contains two parts called reinforcement and matrix. Reinforcement is defined as strong integral and inert component of a composite that is incorporated into the composite matrix to improve its physical properties while the matrix material surrounds and supports the reinforcement materials by maintaining their relative positions [2].

One of high performance fibres applied in composite systems that some research has been conducted to improve its properties is ultrahigh molecular weight polyethylene (UHMWPE). The UHMWPE fibers in general are used in armor, in particular, personal armor and on occasion as vehicle armor, cut-resistant gloves, bow strings, climbing equipment, fishing line, spear lines for spearguns, high-performance sails, suspension lines on sport parachutes and paragliders, rigging in yachting, kites, and kites lines for kites sports [3]. Regarding to those application, at least three main products of UHMWPE-based composite application will be related with this research are air freight containers, portable shields, and wind turbines.

This composite was investigated in the term of mechanical properties with failure mechanism such ILSS test (Interlaminar Shear Strength) to get the higher ILSS value. Failure mechanism is called

also as destructive test include excessive deflection, buckling, ductile fracture, brittle fracture, impact, creep, relaxation, thermal shock, wear, corrosion [4]. ILSS test is a special longitudinal three-point bend test with fibers parallel to the length of the bend bar and the length of the bar being very small [5]. ILSS is determined as most important parameter parameters in determining the ability of a composite to resist delamination damage, which as key parameter for construction material as well.

Andreopoulos, Liolios, and Patriks conducted some similar research but just in mechanical treatment by calendering to eliminate the microfibrillar morphology of the fibre and improve interfacial bonding between fibre/matrix so that better compressive properties can be achieved in reinforced resins [6]. Their research concluded that calendering did not significantly affect fiber strength and only improved adhesive bonding slightly.

Based on the literature above, then this research aimed to improve the ILSS of UHMWPEbased composite by observing some different orientations, calendaring conditions, and optimum glue used either HDPE, LLDPE or reactive glue, then the samples will be evaluated with scanning electron microscope and ILSS tested with failure mechanical tool thus the test results will be vary and the best one could be concluded.

2. Experimental

2.1. Materials

To make a plate of composite, there should be needed reinforcement and matrix/resin. In this study, main reinforce used was UHMWPE tapes (Endumax®) which obtained from Teijin B.V. (The Netherlands), and four of matrixes were HDPE, LLDPE, Epoxy, and Styrene. The calendaring and curing process for HDPE was set at 130ºC while LLDPE was at 120ºC and Epoxy was 70 ºC. Each sheet was stacked until the thickness of approximately 4-5 mm, then pressed/cured in the press. Variables for each treatment were differentiated by temperature, pressure and time. In the other hand, Styrene was also used as alternative matrix. Approximately 95 gram of UHMWPE tape was layered with mixture of 50 grams styrene, DVB, and BPO at room temperature. Then put them in press, temperature was increased slowly to at least 85 °C to allow cross-linking process at 10 bar.

Apart of using HDPE or LLDPE film, epoxy was also used as matrix which applied by using resin and hardener in ratio of 100:34 then rolled the mixture on UHMWPE (66% weight) and pressed on 70ºC 10 bar for 16 hours. Each composite plates then prepared for tests by cutting them in different dimension as requirement of the test with use of Eurolaser® cutter machine.

Table 1. Different practical approach on UHMWPE (Endumax®) tapes by varying the matrix system, pressure and temperature into UHMWPE composite system based on each curing standard

Reinforcement	Matrix/Resin	Pressure	Time
UHMWPE	HDPE	50 bar (130 ºC)	10 min
UHMWPE	LLDPE	50 bar (120ºC)	10 min
UHMWPE	Epoxy	10 bar (80ºC)	16 hours
UHMWPE	Polystyrene	10 bar (85ºC)	1 hour

2.2. Method

The standard reference used for ILSS test was ASTM D2344-48 (Test methods for Apparent Interlaminar Shear Strength of Parallel Fiber Composites by Short-Beam Method). ILSS test was applied by 3-point-bending test method with Instron® load cell of 1kN, 5 kN and 10 kN (span-to-depth ratio was 4:1 and the loading speed was 2 mm/min). The thickness of sample was approximately 5 mm and the width was 9 mm [7]. In other hand, this study also refers to ASTM D790

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(Test methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials). The bending properties test was applied by 3-pointbending test as well with loading speed of 2 mm/min. The thickness of sample was approximately 5 mm and width of 13 mm [8].

The test was continued by observing the samples after ILSS test on Scanning Electron Microscope (SEM). The purpose of SEM analysis was to figure out the failure of bar whether delamination caused by the matrix or reinforce itself. Sample was observed in different scales as well (approximately 2-10μm) and analyzed the failure mechanism before and after bending applied. The equipment used was ΣIGMA® series of Field Emission Scanning Electron Microscopes (FE-SEM) delivers advanced analytical microscopy with the high performance from Carl Zeiss.

3. Results

As aimed in this study, investigating the mechanical properties of UHMWPE composites was conducted by applying different matrixes (HDPE film, LLDPE film, Epoxy resin, Styrene). The mechanical properties included interlaminar shear strength, shear modulus, bending strength and bending modulus are shown below.

Table 2. Mechanical properties of each UHMWPE based composites with comparison of different matrixes

	UHMWPE /HDPE	**UHMWPE/ LLDPE**	**UHMWPE /Styrene**	**UHMWPE /Epoxy**
ILSS (MPa)	5.17 ± 0.04	5.42 ± 0.08	4.82	7.20 ± 0.02
Shear Modulus (MPa)	99 ± 3	148 ± 1	26	200 ± 7
Bending Strength (MPa)	78 ± 1	87 ± 1	55 ± 16	71± 2
Bending Modulus (GPa)	19.7 ± 0.6	19 ± 2	12 ± 2	10.9 ± 0.2
Density (g/cm^3)	0.94	0.96	0.93	0.92

4. Discussion

The first strategy was to apply UHMWPE (Endumax®) as reinforcement, HDPE foil as matrix system. When using UHMWPE and HDPE there was no very significant result, therefore further practical approach was tried by laminating UHMWPE with LLDPE. The reason of choosing LLDPE foil as alternative matrix was because three of them (UHMWPE, HDPE and LLDPE) are from polyethylene but have different properties. The expectation was to see their differences on properties and behavior in the form of composite system.

Based on scanning electron microscope observations, there were some representative images in microscope scale what happened in the bar of UHMWPE/HDPE composite. This scanning electron microscope analysis was done to determine the failure mechanism which indicates shear behavior as well.

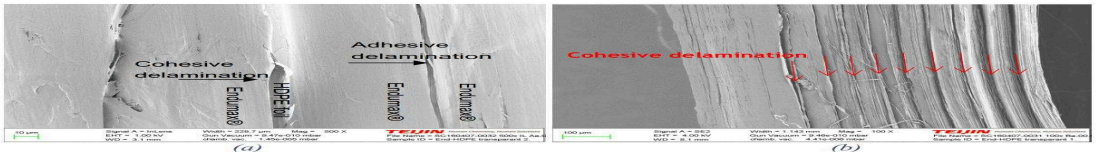

Figure 1. (a) UHMWPE/HDPE appearance on SEM with scale of 10 μm and magnification of 500times shows two types of delamination and (b) scale of 100μm 100 times magnification : the delamination first happened in the interphase of UHMWPE/HDPE then soon followed by delamination within UHMWPE itself

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Based on Figure 1 above, there are two types of delamination/failure on UHMWPE/HDPE composite. The first delamination is called cohesive delamination where the delamination happens between two different material : UHMWPE and HDPE. The second delamination is called adhesive delamination where this failure occurs within one material, in this case is UHMWPE tape. The only delamination that did not occur in this phase is HDPE itself. Thus it could be also concluded that UHMWPE has low adhesion towards HDPE. The conclusion could be first delamination started on HDPE-UHMWPE interphase then soon after that cracked inside the UHMWPE.

The result of ILSS value of LLDPE as matrix was a bit higher than HDPE (see Table 2) where UHMWPE/HDPE was only 5.17 MPa while UHMWPE/LLDPE was 5.42 MPa.

Based on both laminar matrix of HDPE and LLDPE, the expected higher number of ILSS could not be achieved. Then styrene with variable of concentration was expected to give higher strength. The use of styrene applied with DVB and BPO was approximately 20% of UHMWPE sheets weight. Styrene experiment conducted because crosslinked-polystyrene has ever been used in one of the studies as surface modification [9-12]. Therefore there is possibility it could be crosslinked in the UHMWPE which fibrils located; to improve its strength.

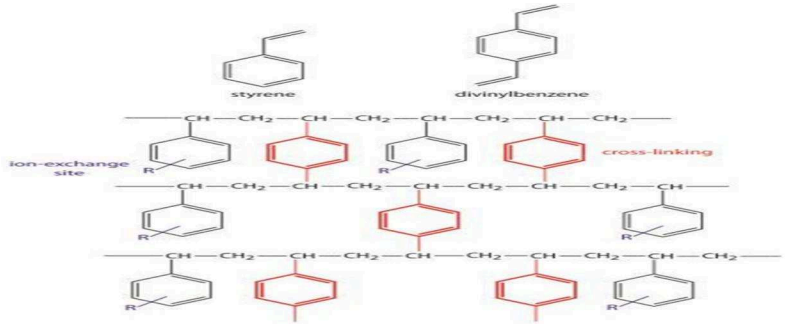

Figure 2. Cross-linking between the styrene-divinylbenzene which later forms poly(styrenedivinylbenzene) resin (-R represents ion exchange site located not R as R branch) [13]

It can be seen from Figure 2 above that reaction of styrene produces linear chain while divinylbenzene cause this styrene chain to be cross-linked. The cross-linking should have enhanced the mechanical stability. This reaction also included participation of free radical chain mechanism which in this case used benzoyl peroxide as radical-producing initiator. When the mixture stirred and heating applied (the press was set at 85°C), the polymerization happened and formed this cross-linked polymer. Resin prepared in this way is called microporous due to its low porosity [14]. This resin is relatively rigid, but unfortunately in this experiment result, the resin could not impact positively to the property of composite because of its low adhesion towards UHMWPE.

Instead of increasing the value, styrene obtained lower value than previous approaches (4.82 MPa of ILSS). During the press, polystyrene was pressed out from the plate. It is possible that just few of them left between the layers. Nevertheless, the samples were brittle but split apart due to low adhesion among the sheets. Thus the experiment was not going to be continued for more polystyrene percentage. Moreover, in scale of electron microscope, we could not see much polystyrene on sample surface due to its low adhesion as mentioned above.

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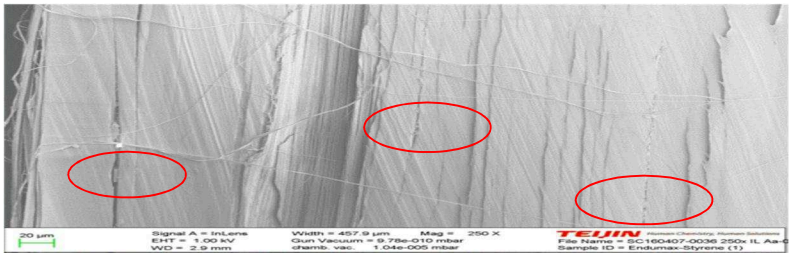

Figure 3. SEM view of UHMWPE/polystyrene composite in the scale of 2μm with magnification 250 times which shows some of polystyrene coming out. It was quiet hard to search the location of styrene because only very few of them appeared

Figure 3 above shows that few spots of polystyrene appeared to the sample surface. The possible reason is because only few styrene left inside the composite due to press and poor adhesion toward UHMWPE. From that figure also we can see that failures happened within

UHMWPE which fibrils of it got loose and delaminated. This indicates UHMWPE obtained its adhesive delamination (failure within Endumax® tape) which this adhesive delamination also happened in previous UHMWPE/HDPE experiment sample. Furthermore, we could see delamination on following figure on the same spot but at higher magnification.

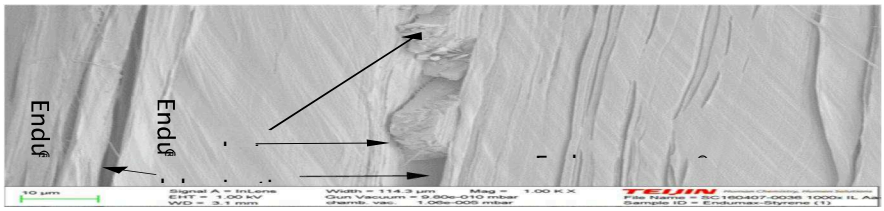

Figure 4. Sample appearance on SEM with scale of 10μm 1000 magnification which shows styrene foam between the UHMWPE (Endumax®) layer where delamination also occurred

It is more obvious from figure above that polystyrene split out of UHMWPE which means the matrix system was failed in this case. When the sample was cut, the fibrils of Endumax was easily fallen apart and it contained polystyrene thus the polystyrene particles were also taken out from the sample.

Due to lower number of strength using HDPE, LLDPE, polystyrene as matrix, another way was introduced by epoxy resin. Epoxy resin is one of the most used matrix/resin in composite system from practical studies, thus it was also chosen to see whether it will give positive influence on UHMWPE composite or not. The experiment was set in 80°C (as the glass transition temperature). In results, the final value of UHMWPE/Epoxy was the highest above all the practical approaches, which was 7.20 MPa of ILSS. The reason behind this phenomena was because of the adhesion of the epoxy was high towards UHMWPE thus the plasticity of the composite increased. The failure mechanism of UHMWPE/Epoxy in microscopic scale is shown in Figure 5 below. Figure 5 shows some delamination between UHMWPE and Epoxy and within UHMWPE itself.

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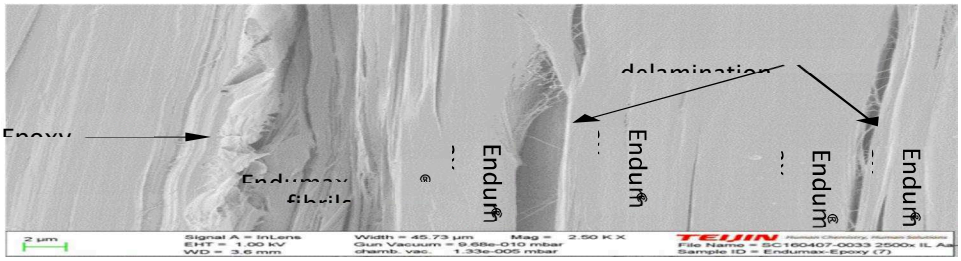

Figure 5. SEM view of UHMWPE which laminated with Epoxy in magnification of 2,500 times with scale of 2 μm, shows some delamination between UHMWPE and Epoxy and within UHMWPE itself

The failure on this composite mostly happened within UHMWPE since adhesion of Epoxy towards UHMWPE was high. This could be shown as Figure 6 below that indicates the epoxy adhesion higher than adhesion of UHMWPE fibrils.

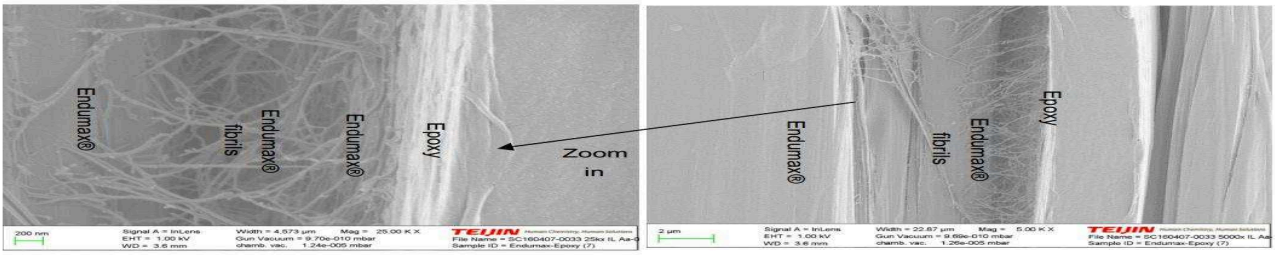

Figure 6. Epoxy on UHMWPE layers view on SEM in scale of 2um and magnification of times and zoomed into magnification of 25,000 times. It shows poor adhesion among fibrils of UHMWPE but better adhesion of Epoxy towards UHMWPE

In conclusion of UHMWPE/Epoxy composite, the adhesion of Epoxy towards UHMWPE (Endumax®) was good as shown on the SEM, while the fibrils of UHMWPE still ripped apart.

5. Conclusion

UHMWPE tapes were treated with different types of matrixes in composite system include HDPE, LLDPE, polystyrene and epoxy, to obtain high value of ILSS and reveal the failure mechanism behind it. The remark from the results of UHMWPE/HDPE and UHMWPE/LLDPE is there was no substantial difference in ILSS. In other hand, polystyrene gave the composite of UHMWPE more brittle properties but split apart due to low adhesion among the sheets thus gave the lowest value of ILSS (4.82 MPa). Above all experiments, the highest number was approached by the UHMWPE/Epoxy (7.20 MPa of ILSS) due to the adhesion of Epoxy towards UHMWPE (Endumax®) was good as shown on the SEM, while the fibrils of UHMWPE still ripped apart. For overall practical approach, the failure mechanism detected in this study was caused by delamination of UHMWPE fibrils. They were ripped apart during mechanical test that lead to delamination of the composite. As summary, in any practical approach conducted with various kinds of matrixes, the UHMWPE composite would keep at low shear strength as long as nothing enhance the interaction between the UHMWPE fibrils.

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