Proceeding Indonesian Textile Conference
(International Conference)
3rd Edition Volume 1 2019
http://itc.stttekstil.ac.id
ISBN : 978-623-91916-0-3

The Effect of Tetraethoxysilane as Filler on UHMWPE (Ultrahigh Molecular Weight Polyethylene)/HDPE Composites

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Abstract : The needed of high performance fibres in technical textile industry is higher from time-to-time since it can replace the heavy metal properties demand. One of popular fibres beside aramid and carbon is called ultrahigh molecular weight polyethylene (UHMWPE) which the high performance polyethylene in numerous research have been observing. Based on some studies, UHMWPE has low adhesion to polymer matrix. Therefore, in this research, to improve the adhesion between UHMWPE fibre and polymer matrix, tetraexthoxysilane was filled into UHMWPE/HDPE surface. The interlaminar shear strength (ILSS) and mechanical properties of the composite were investigated, in comparison with those of the composite without any fillers. In addition, the surface morphology and structure of UHMWPE/HDPE composite filled with TEOS were studied by SEM and EDS. The results showed the ILSS with TEOS improved only 15% from those without fillers but the shear modulus was twice higher. By SEM observation, it showed that UHMWPE/HDPE composite surface was observed with some particles attached. Although there was no chemical interaction between TEOS and UHMWPE/HDPE chains, TEOS has some interaction with UHMWPE molecular chains according to the results of EDS. The TEOS coatings have an effect on the adhesion may be associated with the roughened surface of the composite and upon intermolecular interaction.

Keywords : UHMWPE, HDPE, TEOS, composite, filler, Interlaminar shear
ISBN : 978-623-91916-0-3

1. Introduction

UHMWPE is a linear polyethylene with a molecular weight usually between 3.1 and 5.67 million, and has an average molecular weight more than $2*10^6$ g mol^{-1} [1,2]. Since they have outstanding toughness and chemical resistance, UHMWPE fibres are used in a diverse applications including personal armor, cut-resistant gloves, high-performance sails, artificial joints, and moving parts on weaving machines [3]. In addition, the fibre currently competes with aramid in bulletproof vests, therefore interlaminar shear strength is significantly important as one of main mechanical properties.

UHMWPE fibres have weakness point in wear-resistance and anti-creep properties, therefore, attempts have been conducted by researchers to improve them by filling with in-organic filler, cement, fibre, etc [4-6]. The other approach used is by chemical crosslinking that is introduced by Lewis [7] yet the specific crosslinking using silane was developed since 1970s in insulation materials of high voltage electric cables [8]. Another study by Atkinson and Cicek [9] was also conducted where the silane crosslinked polyethylene for orthopedic applications. However, only slight information is available on silane crosslinked UHMWPE, which one of them is the study of Sung In Moon [10], that applied organo alkoxysilane (⊚-methylmethacryloxypropyltrimethoxysilane) where it could improve ILSS of UHMWPE from 8 to 16 MPa. Therefore, in this study, UHMWPE was tried to be crosslinked by other silane called tetraethoxysilane with the aim of improving its mechanical and interlaminar shear properties.

2. Materials and Methods

To make a plate of composite, reinforcement and matrix/resin should be needed. In this study, main reinforce used was UHMWPE tapes (Endumax®) which obtained from Teijin B.V. (The Netherlands), and the matrix was HDPE. The calendaring and curing process for HDPE was set at 130ºC 50 bar for 10 minutes. 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. Practical approach was applied by using a filler called TEOS which was set on 88% weight of previous calendared UHMWPE/HDPE sheets by immersing the sheets in TEOS in vacuum desiccator for total of 70 hours and drying the sheets at 80ºC for 24 hours. The curing process was set on 50 bar at 130 ºC for 10 minutes. Each composite plates then prepared for tests by cutting them in different dimension as requirement of the test with use of Eurolaser® cutter machine.

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 [11]. On the other hand, this study also refers to ASTM D790 (Test methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials). The bending properties test was applied by 3-point-bending test as well with loading speed of 2 mm/min. The thickness of sample was approximately 5 mm and width of 13 mm. [12].

3. Results

As aimed in this research, to improve the adhesion between UHMWPE fibre and polymer matrix, tetraexthoxysilane was filled into UHMWPE/HDPE surface. The mechanical properties of the composite were investigated, in comparison with those of the composite without any fillers. The mechanical properties included interlaminar shear strength, shear modulus, bending strength and bending modulus are shown below.

Table 1. Mechanical test results of ILSS test on UHMWPE based composites

Mechanical Properties	UHMWPE/HDPE	UHMWPE/HDPE+TEOS
ILSS (MPa)	5.17 ± 0.04	5.98 ± 0.06
Shear Modulus (MPa)	99 ± 3	184 ± 7
Bending Strength (MPa)	78 ± 1	55 ± 5
Bending Modulus (GPa)	19.7 ± 0.6	41 ± 1
Density (g/cm^3)	0.94	0.94

ISBN : 978-623-91916-0-3
DOI : 10.5281/zenodo.3470990

4. Discussion

The chosen filler applied in this research was TEOS that aimed to get silica layer into the composite and expected to higher the mechanical properties. The concentration of TEOS was 88% of UHMWPE/HDPE sheets in total weight. This concentration was chosen because TEOS (liquid form) should get into all pores inside the UHMWPE sheets in immersing box (sheets thickness was about 4 mm). Therefore, the setting also took the advantage of vacuum and desiccator to allow silane getting into the pores of UHMWPE. The curing processes of both composite samples (with and without TEOS) were same, 50 bar for 10 minutes at 130ºC. There was no other heating process after the curing, but the cooling process took place at room temperature inside the laboratory cabinet to stabilize both composites before mechanical tests.

The mechanical properties of two composites before and after adding filler (TEOS) for 24 hours are presented in Table 1. It can be seen that the result on Table 1 shows that TEOS gave partial positive influence on ILSS by increasing its value with the same pressure, temperature and time condition; from 5.17 MPa to 5.98 MPa that approximately 15% higher. Meanwhile, the shear modulus and bending modulus were twice higher from 99 MPa to 184 MPa and 19.7 to 41 GPa, respectively. This means that the treatments by adding filler have an no significant effect on ILSS but play a role in increasing the bending modulus of the composites.

Besides the mechanical properties shown on Table 1, the stress-strain curve was also given below in Figure 1. The curve indicates whether the composites are ductile or brittle. The blue line represents UHMWPE/HDPE composite that shows the material has lower stress but higher strain which also means it is ductile and has characteristic of plastic but large toughness since it absorbs more energy under the curve.

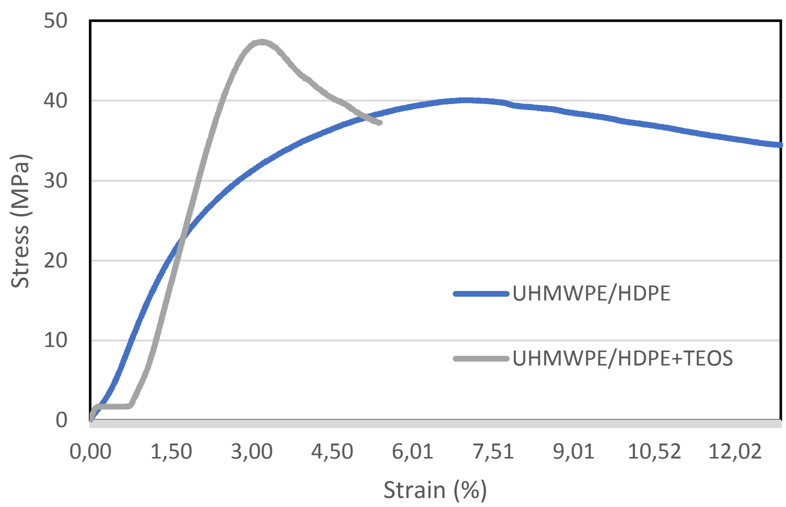

Figure 1. Stress-strain curve of UHMWPE/HDPE composites with and without TEOS shows the elasticity of the composites where the blue line represents UHMWPE/HDPE composite (more ductile) while the grey line represents UHMWPE/HDPE/TEOS composite (more brittle)

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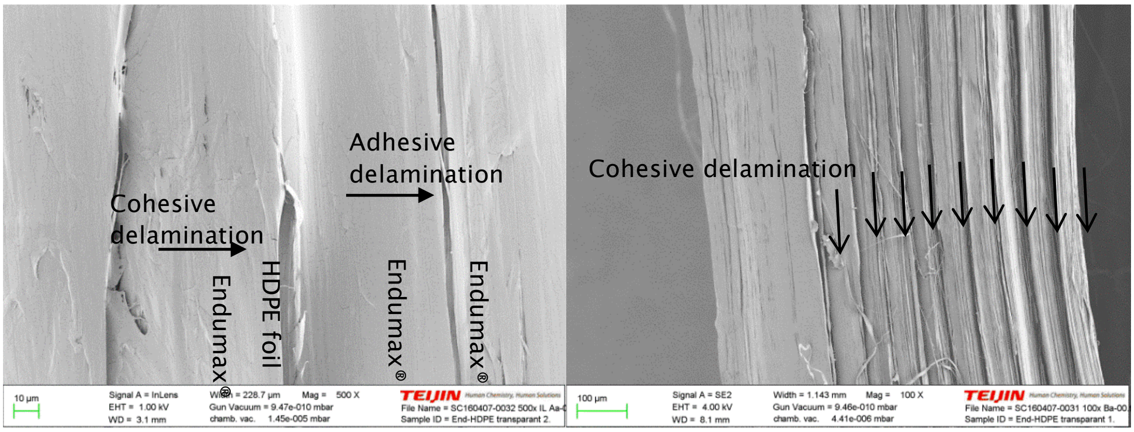
On the other hand, the grey line represents UHMWPE/HDPE composites which treated with TEOS showing a higher peak and stress but lower strain. This means the adding of TEOS significantly gives impact to the composite in term of elasticity. By adding TEOS, the composites become brittle and has smaller toughness because it absorbs less energy under the curve. Moreover, the relation between Table 1 and Figure 1 can explain that number of bending and shear modulus (Table 1) are twice higher indicates the increase of stiffness (brittle) and strength of the material which presents with higher peak of UHMWPE/HDPE/TEOS in stress-strain curve.

Moreover, there are some representative pictures in microscope scale revealed 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.



(a) (b)

Figure 2. (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

Based on Figure 2 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 materials: 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.

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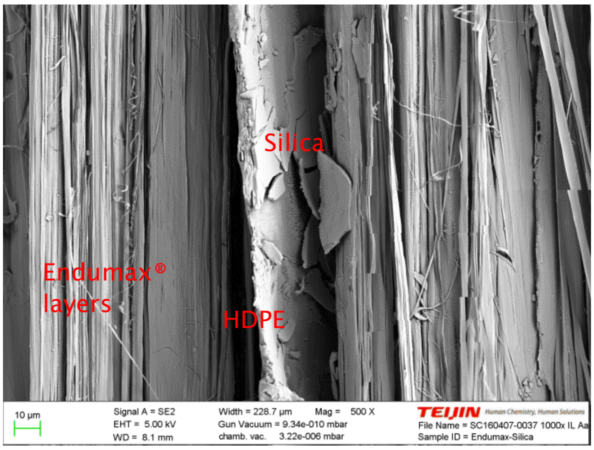

Figure 3. Silica was shown as cracked part on spot between HDPE and Endumax (scale 10µm magnification of 500 times**)**

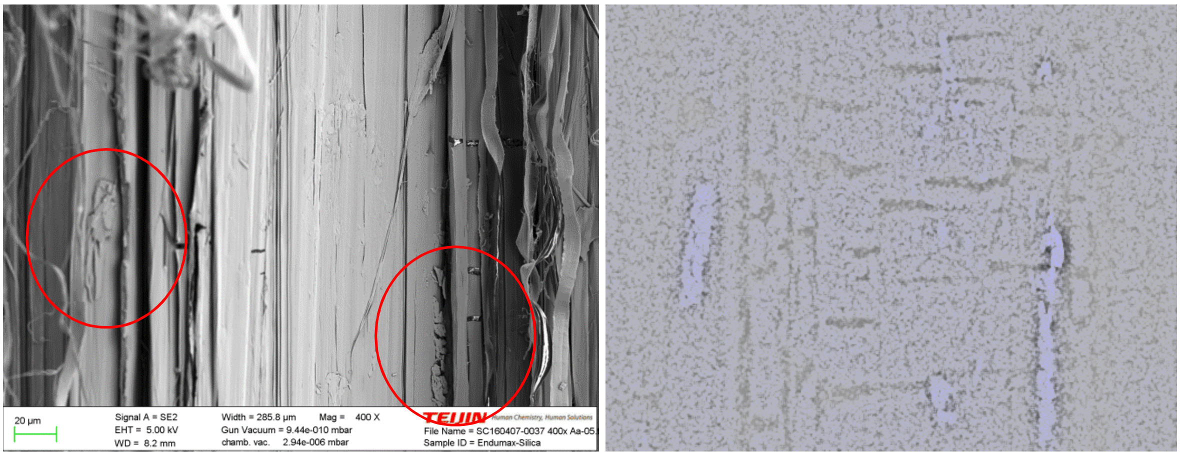

Figure 4. Silica was found in some spots in the interphase. It was shown by SEM (left) and while the right figure is by EDS mapping to read signal of Si (Silica) from the same spots.

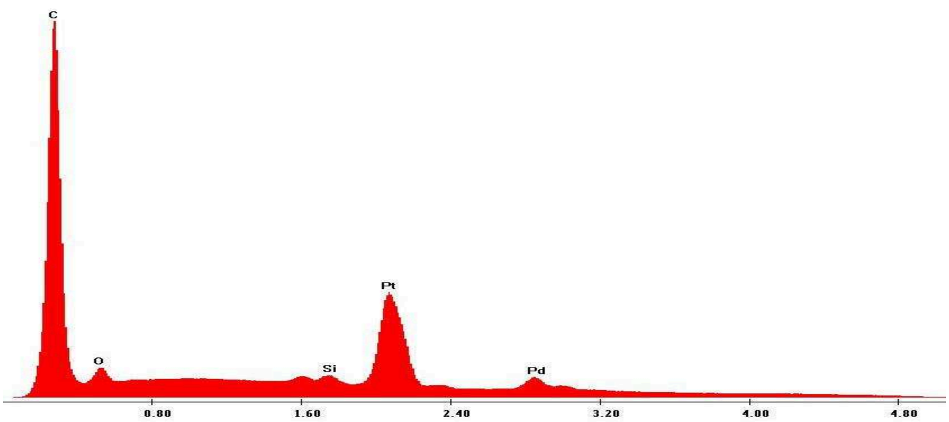

Figure 5. EDS mapping of the components from Figure 4 where the Si (Silica) element concentration was present. The other components (C,O) were part of the polyethylene composite while Pt and Pd was used as coating for microscope purpose

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On the other words, Silica was barely hard to find from layers in between UHMWPE/HDPE on SEM analysis (Figure 3). The silica was found cracked and it was on low silica concentration by observing on EDS mapping in the same spot as it found (Figure 4 and 5). As obtained from previous SEM of only UHMWPE/HDPE, here delamination also occurred on interphase of UHMWPE/HDPE and within UHMWPE (cohesive and adhesive delamination). The silica kept outside the layer instead of getting into pores. On EDS mapping on figure 5 presents the percentage of silica was low which indicates TEOS was not absorbed enough into the layers of UHMWPE/HDPE composite.

5. Conclusion

In order to improve the interlaminar shear strength of UHMWPE fiber-reinforced polymer matrix composites, specifically UHMWPE/HDPE composite, filler was added to the system, named tetraethoxysilane (TEOS) which has been observed. According to the above analysis, the difference between UHMWPE/HDPE composite with UHMWPE/HDPE filled TEOS was not significant in term of interlaminar shear behaviour. The silica layer on UHMWPE/HDPE may increase the interfacial adhesion of UHMWPE/HDPE composites whose interface did not posses a chemical binding, but the higher ILSS could be achieved even only 15%. This means the interlaminar between UHMPWE fibres and HDPE in composite system obtained higher strength physically not chemically since the TEOS could not be filled into the pores of the fibre. In term of elasticity, by adding TEOS, the UHMWPE/HDPE composites became more brittle and absorbed more energy under the curve of stress-strain. The significant higher value of ILSS was not obtained, however, we knew the failure mechanism by experiments: the weak part of UHMWPE was in UHMWPE tape fibrils inside the composite, even adding the filler (TEOS) did not improve the fibrils properties. Therefore, for further experiment, the focus should not only be targetted on surface treatment, but also more onto the fibrils level area such as x-ray treatment or practical approach in molecular area. Then this result will be expected to higher the strength of UHMWPE based composite which later could be applied in future technical textile production.

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