

## EFFECT OF CELLULOSE-BASED FIBERS EXTRACTED FROM PINEAPPLE (ANANAS COMOSUS) LEAF IN THE FORMATION OF POLYURETHANE FOAM

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### ABSTRACT

New polyurethane foams were fabricated utilizing cellulose-based fibers extracted from pineapple (*Ananas comosus*) leaf as raw material. The pineapple leaf fibers (PALF) were treated with alkali and subsequently bleached to enhance its fiber-matrix adhesion. Polyurethane composites have been prepared by incorporating 10% cellulose-based fibers extracted from PALF during polyurethane synthesis. The Fourier transform infrared (FTIR) spectra revealed that increase in the C-H and C-O vibrational modes absorption were observed when cellulose-based fibers were incorporated which might be attributed to the successful bonding of high purity cellulose-based fibers in the matrix of polyurethane foam. The physico-chemical, spectral and thermal properties of the polyurethane foam and the cellulose-based fibers reinforced polyurethane composites have been studied.

**Keywords:** Cellulose-based fibers; Pineapple leaf fibers; Polyurethane foam

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## 1. INTRODUCTION

Polyurethane (PU) has been introduced from the year 1950's and its attention widely increases after the introduction of plastic foams. PU belongs to a wide-range and highly diverse family of polymers with varied properties such as solid and open cellular structures. Traditionally, it has been widely used in furniture, textiles products, footwear, packaging, bed mattresses, carpet, cushions and elastomers. Current utilization of PU composites include aerospace, marine and automobile applications [1-3]. PU foam is normally synthesized by mixing the basic components which include isocyanate, polyols, surfactants and catalysts [4].

Polyols are the main source of hydroxyl or other isocyanate reacting groups. The group of Y. Li tried to use fatty acids, fatty acid esters and crude glycerol as a source of polyols. Furthermore, crude glycerol is a very promising renewable feedstock for polyols and PU. On the other hand, it is difficult to obtain polyols with consistent quality and properties when using crude glycerol due to its varied composition [5]. The depletion of the petroleum-based crude oil reserves, give way to the utilization of soybean- based derived polyols as essential sustainable alternative for petroleum-based materials to ensure longevity of the product [6]. Zhang et al. synthesized epoxidized soybean oil (ESBO) polyols as replacement to the conventional petroleum-based polyols. Their result shows that the petroleum-based polyols exhibit a lesser tensile strength as compared to ESBO polyols. Furthermore, the Young's modulus of PU foam utilizing ESBO polyols significantly enhance suggesting that the mechanical property of the produced PU foam improves [7].

To improve the PU foam properties further, researchers tried to synthesize PU foam using fillers or additives for the enhancement and modification of the material properties. The addition of fillers in the PU foam increases the density and resistance in terms of the compressive strength of the material. However, fillers reduce the resilience and contribute to the increase in the permanent deformation. Proper optimization of the filler is needed to obtain reliable quality of the final PU foam. Previous works used calcium carbonate, silica, carbon black and nano-silica as fillers to the PU foam formation. However their results revealed that less mechanical enhancement on the foam was observed [8-11]. Researchers nowadays tried to synthesize PU foam with the addition of natural fibers as filler and reinforcement. The utilization of natural fibers as reinforcement to PU foam shows that the

Young's modulus and flexural strength increases as they increase the fiber content [12,13].

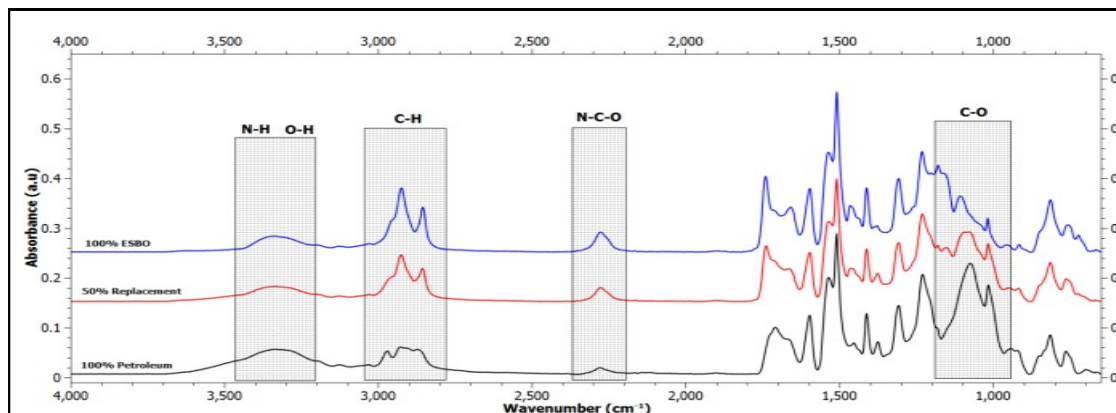
One of the promising natural fibers which are renewable and low production cost is coming from pineapple leaf. Pineapples are very common plants in tropical countries [14]. In fact, in the year 2002 to 2012, worldwide pineapple production was gradually increasing and Philippines is one of the leading countries which produces pineapples world wide [15].

These kind of pineapple production in the Philippines constitute a large amount of agricultural wastes since industries utilized only the pineapple fruit for food production while the pineapple leaves are considered agricultural wastes. In fact, some researchers utilized pineapple leaf fiber (PALF) in textile purposes and paper production [16]. On the other hand, the use of PALF as reinforcement material for epoxy, polyethylene, polypropylene, vinyl ester, polyester, polycarbonate and low density polyethylene base PALF composite have been successfully explored [17]. However, there have been no reports on the utilization of PALF as reinforcement to the polyurethane foam production. This paper will elucidate the effect of cellulose-based fibers extracted from PALF for polyurethane foam formation.

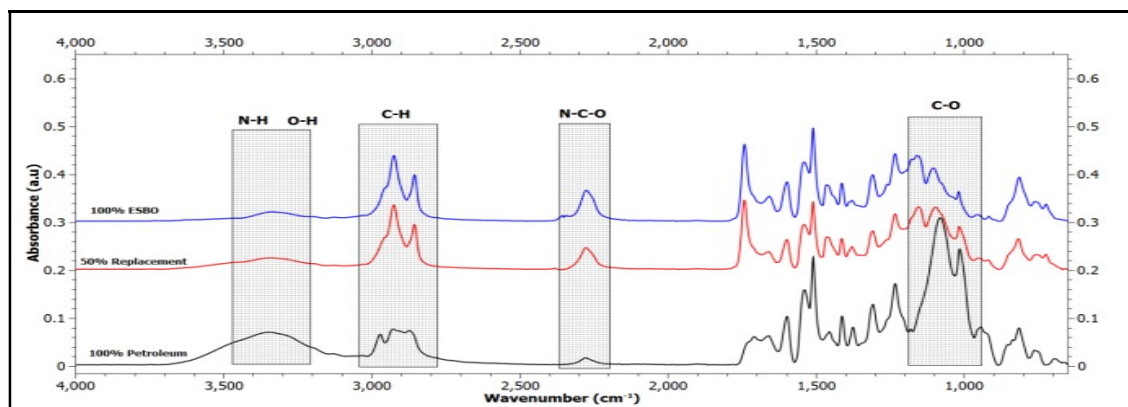
## 2. RESULTS AND DISCUSSION

Figure 1 shows the the FTIR spectra of the polyurethane foams without incorporation of cellulose-based fibers while Figure 2 shows the FTIR spectra of polyurethane foams with 10% incorporation of cellulose-based fibers extracted from pineapple (*Ananas comosus*) leaf (PALF). The broad peak around 3410  $\text{cm}^{-1}$  belongs to the hydroxyl group which corresponds to the intermolecular and intramolecular H-bond of the free OH- of cellulose from PALF. This may include the vibrational mode coming from the N-H bond of the polyurethane matrix. The overlapping and the vibrational stretching of O-H and N-H as shown in Figures 1 and 2 may be associated to the urethane linkages with the hydrogen bonds. It is expected that the stoichiometric ratio of the isocyanate (NCO) and the polyols (OH-) should be maintained at unity to preserve the good quality of the polyurethane foam. However, the drastic decrease of O-H in the absorption spectra of the PU foam with 10% cellulose-based fibers may trigger ruptures on the cell wall structure of the PU foam since the amount of O-H affects the cell wall structure of the PU foam. This might be due to the

insufficient number of O-H present in the bleached treated PALF that would trigger the stoichiometric ratio becomes far from unity.



**Fig.1.** FTIR spectra of polyurethane foams without cellulose-based fibers.

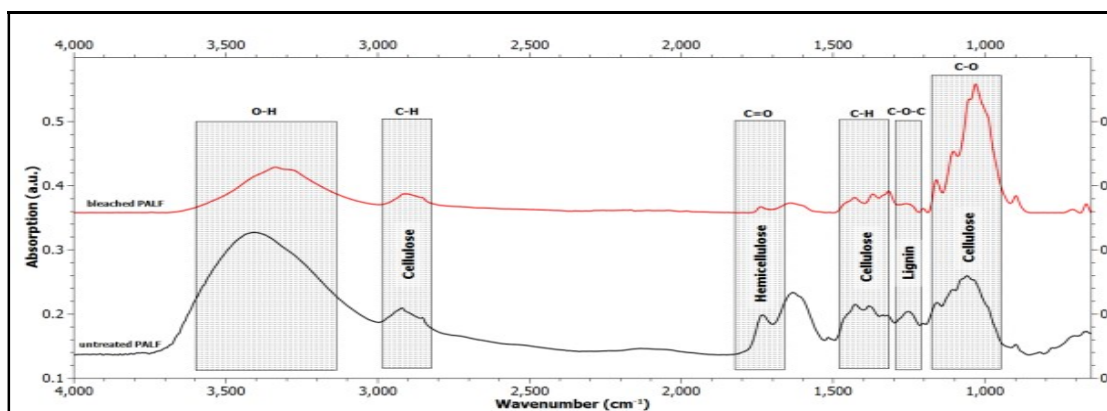


**Fig.2.** FTIR spectra of polyurethane foams with cellulose-based fibers.

The broad peak at around 2921-2852  $\text{cm}^{-1}$  is associated to the C-H asymmetric stretching while the peak around 1100  $\text{cm}^{-1}$  corresponds to the C-O asymmetric stretching for the polyurethane foams with and without 10% incorporation of cellulose-based fibers. The significant increase of absorbance corresponding to C-H and C-O asymmetric stretching were observed when 10% cellulose-based fibers were added during the PU foam formation. The increase of the absorbance in the C-H peak for the PU foam with 10% cellulose-based fibers can be attributed to the presence of cellulose in the bleached treated PALF. This means that the cellulose-based fibers present in the bleach treated PALF successfully bonded to the polyurethane foam during the formation process. On the other hand, it can be easily noted

that by increasing the bio-based polyol replacement, there is a significant increase of the vibrational mode around  $2290\text{ cm}^{-1}$  corresponding to the NCO peak. Furthermore, incorporating 10% cellulose-based fibers into the PU matrix also contributes to the increase of NCO peak at around  $2290\text{ cm}^{-1}$ . This may be due to some unreacted NCO present in isocyanate.

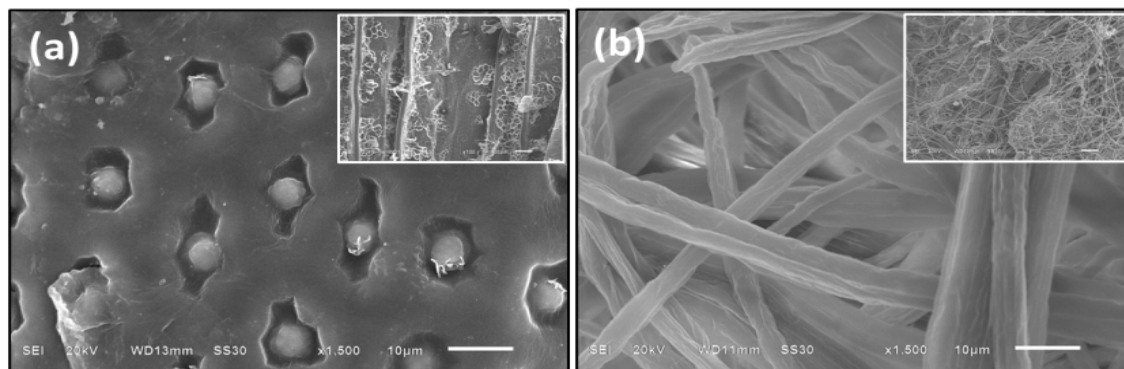
In order to determine if we successfully extracted cellulose-based fibers from pineapple leaf, we performed the FTIR spectroscopy of the untreated and bleached treated PALF. Figure 3 shows the FTIR spectra of the untreated and bleached treated pineapple leaf fibers. It is noted that bleaching the pineapple leaf will subsequently produce a high quality cellulose-based fibers. The FTIR spectra of the bleached treated PALF will reduce the absorbance coming from hemicellulose, lignen and other impurities. The absorbance around  $1050\text{ cm}^{-1}$  corresponds to cellulose which significantly increase after bleached treatment. This implies that high quality cellulose-based fibers are produced after bleached treatment.



**Fig.3.** FTIR spectra of the untreated and bleached treated pineapple leaf fibers.

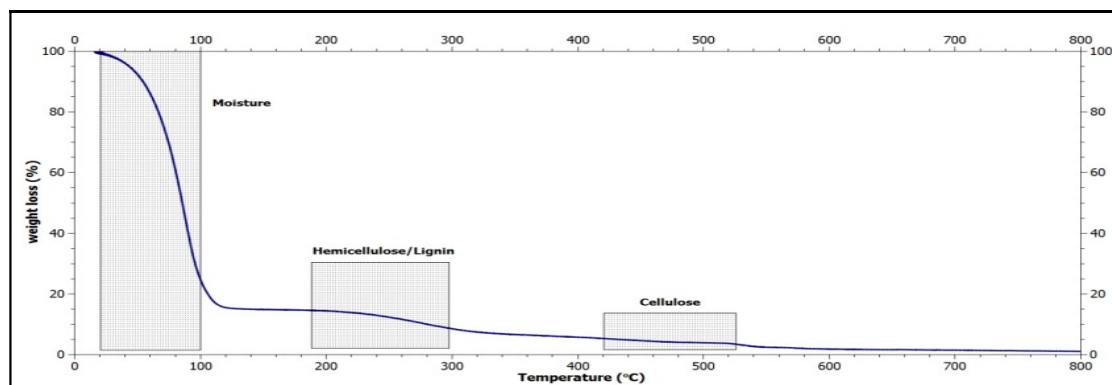
The Scanning Electron Microscopy (SEM) images revealing the surface morphology of the untreated and bleached treated pineapple leaf fibers are shown in Figure 4. It is observed that high quality cellulose-based fibers are significantly produced after bleached treatment. A well-defined pineapple fibers are very apparent in the SEM image as shown in Figure 4(b). On the other hand, a cement-like structures are present in the untreated pineapple leaf fibers. This may be composed of hemicellulose, lignen and other impurities that was also observed in the FTIR spectra as shown in Figure 3. The removal of hemicellulose, lignen and other

impurities on the surface of the PALF resulted to higher fibrillation of the PALF. Basically, surface treatment of the PALF could improve the surface wettability of the fiber. The high quality surface of bleached treated PALF shows a good matrix-fiber adhesion if incorporated into the formation of PU foam. It is very apparent that bleached treatment significantly produce a high quality cellulose-based fibers from pineapple leaf.



**Fig.4.** SEM images of the (a) untreated (b) bleached treated pineapple leaf fibers. Inset photos are the lower magnification of the SEM images of both samples.

The Thermal Gravimetric Analyzer (TGA) spectra of the untreated PALF is shown in Figure 5. The decomposition is composed of a three-step process. The first step process was shown at temperature between 20.54°C to 100°C and had a weight loss of 83.7% which constitute to the degradation of water moisture present in the sample. While the second step process was shown at temperature from 215°C up to 324°C with a weight loss of 6.69% which found to be the degradation temperature of hemicellulose. In this stage, cellulose-based fibers remained in the structure. The third step process was shown at 518°C to 542°C and had a weight loss of 1.15% where cellulose-based fibers might be degraded.



**Fig.5.** TGA spectrum for the untreated pineapple leaf fibers.

### 3. EXPERIMENTAL

Pineapple leaves were washed with water and air-dried for three days, cut into small pieces and grind using blender. Alkali treatment was done by mixing the PALF with sodium hydroxide (NaOH) aqueous solution under mechanical stirring for about 4 hours at 100°C. Alkali treatment using NaOH was used to remove hemicellulose, lignin and other soluble materials. The alkali treated PALF was washed several times with distilled water. Furthermore, alkali treated PALF then undergo bleach treatment using sodium chlorite (NaClO<sub>2</sub>) aqueous solution for 4 hours at 100°C. Bleached treated PALF was washed several times with distilled water and dried.

The production of PU foam was done using free-rise method. This was done by mixing A-side and B-side components. The A-side component was composed of PAPI 27 isocyanate (polymeric MDI) while the B-side component is composed of Epoxidized soybean oil (ESBO) and petroleum-based polyols (VERANOL™ 490), Polycat 5 and Polycat 8 as blowing catalyst, Dabco DC 5357 as surfactant and distilled water as blowing agent. B-side components were mixed utilizing 10% of bleached treated PALF per unit volume of the B-side component. Polyurethane foam formation is regularly done by mixing polyols, surfactant and catalysts to create faster foam formation. For the B-side component, 10% of the bio-composites PALF per unit volume of the total polyols (B-side) were mixed to the solution of ESBO, catalysts, surfactants and blowing agent. The solution was then mixed with the VORANOL™ 490 to complete the B-side component which constitutes the polyols. The process of mixing A-side and B-side components utilizing the 10% cellulose-based PALF was done with constant stirring to obtain homogenous solution at room temperature for about 10-15 seconds and allow to rise freely. The formation of PU foam was done by continuously mixing the B-side components which consists of VORANOL™ 490, ESBO, PALF, Polycat 5, Polycat 8, Dabco DC 5357 and distilled water as the blowing agent. After mixing the B-side components, it was mixed with the A-side component consist of isocyanate.

The functional groups and vibrational modes of the untreated and bleached treated PALF and the PU foam with and without 10% cellulose-based fibers were determined using Fourier transform infrared spectroscopy (FTIR). The surface morphology were determined using scanning electron microscope (SEM) of both untreated and bleached treated PALF. Thermal

decomposition and the weight loss of untreated PALF was done using thermogravimetric analysis (TGA).

#### 4. CONCLUSION

The effect of incorporating cellulose-based fibers extracted from pineapple (*Ananas comosus*) leaf as raw material in the formation of polyurethane foam was investigated. The formation of PU foam with 10% cellulose-based fibers significantly increase the C-H and C-O vibrational modes that can be attributed to the successful bonding of high quality cellulose-based fibers with the polyurethane foam during the formation process. Bleached treatment of the pineapple leaf fibers will produce a high quality cellulose-based fibers with lesser amount of impurities. This is a promising route towards the production of bio-based polyurethane foams.

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