Structural and Dielectric Properties of Polyurethane Palm Oil Based Filled Empty Fruit Bunch

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Abstract— *This paper presents the structural and dielectric properties of polyurethane palm oil based filled empty fruit bunch. Polyurethane filled with empty fruit bunch (PU-EFB) composite was prepared through hot press process. The crystal structure of PU-EFB has been studied using X-ray diffraction technique. The effect of EFB filler content on the microwave dielectric properties of the PU-EFB composites was studied in the X-Band frequency region using Vector Network Analyzerat room temperature. A Kraszewski model showed good correspondence with measured dielectric constant of PU-EFB composite, especially at the higher loading level.*

Keywords—dielectric properties, PU-EFB composite, polyurethane palm oil based.

I. INTRODUCTION

With the rapid development of modern microwave communication systems, microwave dielectric ceramics have attracted much scientific and commercial attention. The main demand for technology is the use of low cost and produce high-performance product. Dielectric materials play an important role in leading this industry. A small ceramic component made from a dielectric material is fundamental to the operation of filters and oscillators in several microwave systems[1]. The selections of dielectric samples are a major problem in fabricating microwave substrate[2].

The process to produce microwave substrate with high performance gives an impact to the environment. Most of them are thermoset resin-based systems[2].High cost is one of the problems in bringing high frequency personal communication equipment to the consumer market. Materials that provide superior microwave performance for the realization of active devices and passive elements are typically very expensive[3].

Polyurethane (PU) plays an important role in modern life and its global demand is growing every year. To date, there are many researches on renewable resources for polyurethane. It has been reported that polyurethanes are a polymer that can be prepared using natural product such as palm oil[4].

This research presented the designing of microwave substrate using palm oil based polyurethane and empty fruit bunch (EFB) as filler. EFB is a waste material from palm tree. The ingredients used in the processes of preparing oil based polyurethane were ingredients used in the manufacturing of polyurethane, namely *p*-MDI and palm oil based glycerol. The polyurethane-EFB (PU-EFB) composite was prepared by using hot press method. The structural characterization of PU-EFB composite and the effect of filler loading on dielectric properties were reported.

II. EXPERIMENT

2.1 Material

The raw materials used in this study were glycerol palm oil based with molecular weight 92.09380which was obtained from FPG OleochemicalSdn. Bhd. Kuantan, Pahang and Diphenylmethane-4, 4’-diisocyanate (p-MDI), Sigma-Aldrich with molecular weight: 250.25200. The empty fruit bunch was obtained from Kilang Kelapa Sawit Felda Neram, Kemaman, Terengganu.

2.2 Preparation of empty fruit bunch powder

The obtained EFB fibre was air-dried for 1 week before being ground. The ground EFB was then sieved using 70 µm a sifterto separate the powder to different particle sizes. Soaked using distilled water for24 hours to remove ash[5]. The EFB then dried at 90ºC until the mass was constant.

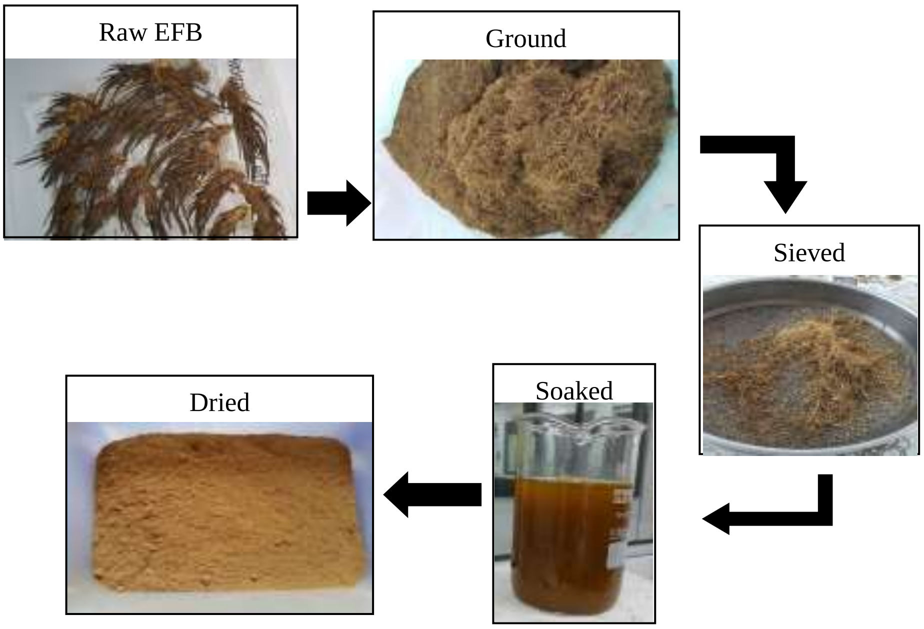

Fig.1: Preparation of empty fruit bunch powder

2.3 Preparation of polyurethane using glycerol palm oil based

The formation of polyurethane foam in this study resulted from the exothermic reaction between glycerol palm oil based and Diphenylmethane-4,4'-diisocyanate (*p*-MDI). Figure 2 represented the chemical reaction between glycerol palm oil based and isocyanates.[2]

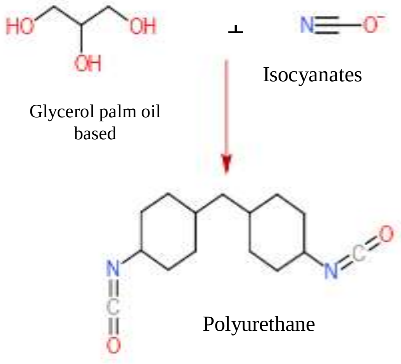

Fig.2: Chemical reaction of polyurethane

The amount of glycerol and *p*-MDI were fixed at 3g and 6g respectively with ratio 1:0.5[6]. Glycerol palm oil based was first weighed in a beaker and gently stir 5 minutes. The *p*-MDI mixed and stirred vigorously for 10 seconds, poured in aluminium for self-rising. The foam was conditioned at room temperature for at least 24 hours and preceded with curing process until the mass of the polyurethane was constant.

2.4 Preparation of PU-EFB composite

Figure 3 shows the process to prepare PU-EFB composite. Polyurethane was finely ground and mixed with EFB powder. The powder mixture was mixed thoroughly within 30 seconds and was put in a mould the size of 22.86 x 10.16 mm (x-band dimension mould size). The PU-EFB compositewas heatedat 40 ^{o}C and 20 kgcm^{-2}with hotpress methods within 15 minutes. PU-EFB composite sheet was cooled for 1 hour and then removed from the mould. The curing process was carried out by heating PU-EFB composite sheets at a temperature of 70^{o}Cand placed in a vacuum pump until they reached constant mass.The composites were polished to ensure a good quality surface and parallel surfaces.

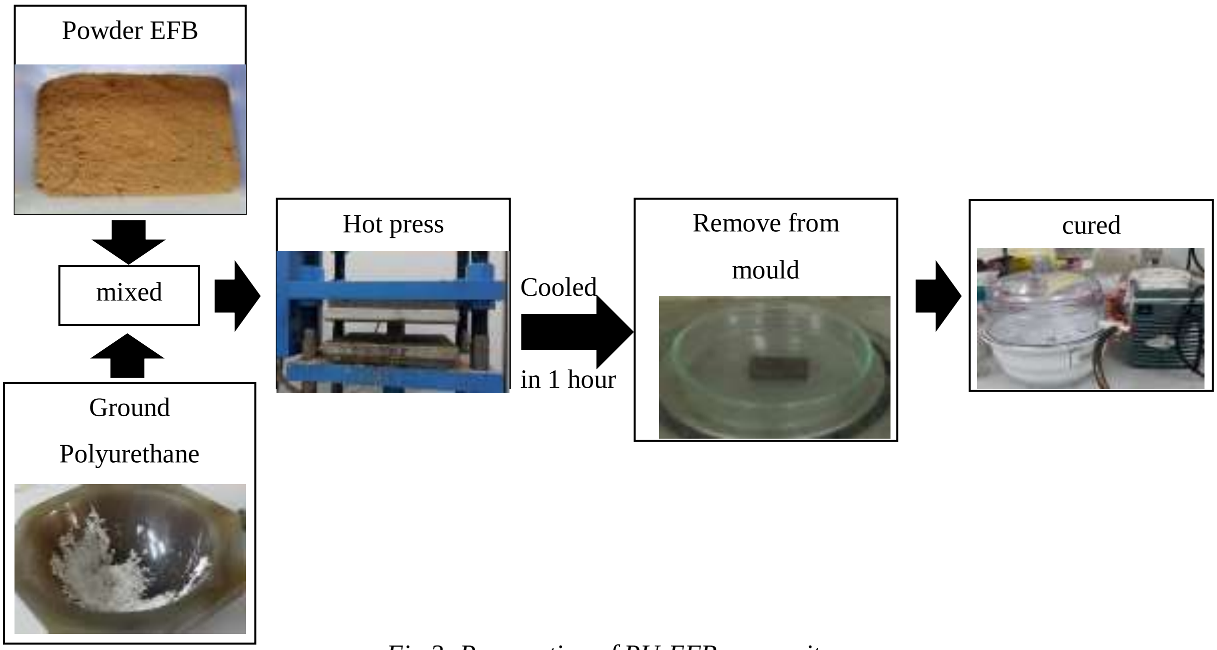

Fig.3: Preparation of PU-EFB composite

III. RESULTS AND DISCUSSION

3.1 Structural Properties

The structural of crystallinity of PU-EFB composite was studied using X-ray diffraction technique at room temperature with monochromatic Cu Kα (λ=1.540562Å) in the scan range of 10° and 80°. Figure 2 shows the X-ray diffraction pattern of 0% EFB (pure PU), 40% EFB, 60% EFB and 80% EFB. The powder diffractograms from X-ray diffraction exhibits broad peaks at 2θ angles around 22.72°, 18.86° and 41.5°.These indicate that some degree of crystallinity of polyurethane.

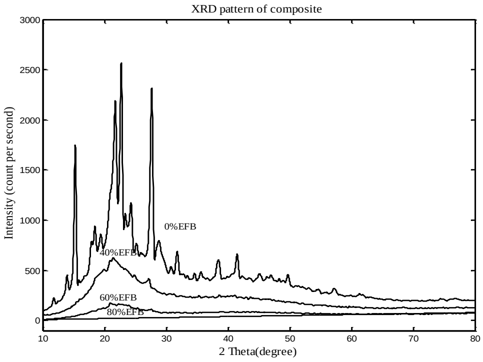

Fig.4: XRD pattern of composite

The graph also shows a decrease in the peak as a percentage of filler loading that entered into the PU matrix increased. At 80% filler loading, it was found that almost no peak occurred. This phenomenon concluded that the crystallinity of the composite was controlled by the presence of filler loading percentage in the composite.

3.2 Dielectric Properties

The dielectric properties of composite were studied using Agilent E5071C Network Analyzer in conjunction with Agilent 85070E dielectric probe. The dielectric measurement was conducted over a frequency range from 8 GHz to 12 GHz (X-band) at room temperature. Figure 2shows the variation of dielectric constant for composite at x-band frequency. The value for polyurethane (0% EFB) was between 2.5 to 2.6. The dielectric constant had slightly decreased when polyurethane was mixed with the EFB. Overall, the value for dielectric constant for composite was within the range of 2.1- 2.5.

Crystallinity of the composite effects the value of dielectric constant. When the crystalline was at high value, the dielectric constant was higher than when the structure of the composite was amorphous. The optimum value for dielectric constant was obtained when the composite contained 40% EFB.

The loss factor for polyurethane (0% EFB) was high compared to loss factor of the composites. The loss factor of each composite was low and it approached the value zero as shown in figure 4. At the frequency of 9.8GHz to 10.3GHz it slightly increased the value of loss factor.

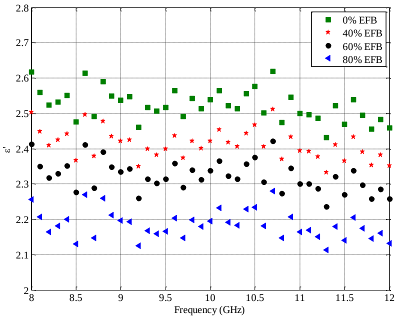

Fig.5: Relationship between dielectric constant and frequency for different percentage of EFB.

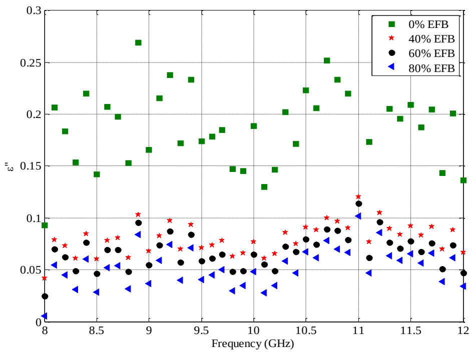

Fig.6: Relationship between loss factor and frequency

Variation of dielectric constant and loss factor as a function of percentage EFB loading in polyurethane is shown in figure 7a to figure 7e. For dielectric constant at 8 GHz, the gradient at 0.4 % to 0.6 % was –0.45 and the gradient at 0.6 % to 0.8 % was -0.078. For dielectric constant at 9 GHz, the gradient at 0.4 % to 0.6 % was -0.043 and the gradient at 0.6 % to 0.8 % was-0.069. For dielectric constant at 10 GHz, the gradient at 0.4 % to 0.6 % was --0.0418 and the gradient at 0.6 % to 0.8 % was-0.0713. For dielectric constant at 11 GHz, the gradient at 0.4 % to 0.6 % was -0.0468 and the gradient at 0.6 % to 0.8 % was-0.0685. For dielectric constant at 12 GHz, the gradient at 0.4 % to 0.6 % was -0.0468 and the gradient at 0.6 % to 0.8 % was -0.0631.For loss factor at 8 GHz, the gradient at 0.4 % to 0.6 % was -0.0087 and the gradient at 0.6 % to 0.8 % was -0.0095 .

For loss factor at 9 GHz, the gradient at 0.4 % to 0.6 % was -0.0067 and the gradient at 0.6 % to 0.8 % was-0.009. For loss factor at 10 GHz, the gradient at 0.4 % to 0.6 % was -0.0062 and the gradient at 0.6 % to 0.8 % was-0.0083. For loss factor at 11 GHz, the gradient at 0.4 % to 0.6 % was -0.003 and the gradient at 0.6 % to 0.8 % was-0.0062. For loss factor at 12 GHz, the gradient at 0.4 % to 0.6 % was -0.0097 and the gradient at 0.6 % to 0.8 % was-0.0064. From the trend of the gradient of dielectric constant and loss factor graph, the graph had decreased linearly at range of 0.4 % to 0.6 % and 0.6 % to 0.8 %. At range of 0.4 % to 0.6 % the gradient is lower than the slope of the graph at range 0.6 % to 0.8 %. At low filler loading, the dielectric constant high, where at this point the composite was introduced to the PU matri**x.**

PU filled EFB composite exhibited stable dielectric properties with respect to frequency and minimum dielectric anisotropy. There were some defects such as air gap, water and the interface phase between EFB and PU in composites materials, which can influence the relative dielectric constant and the dielectric loss of the composites.

Using hot press technique in production of the substrate composite, the interface region occurred between the filler and matrix. At higher filler loading, where a high amount of EFB, the interface region was reduced in between the filler and matrix and this couldinfluence the dielectric constant and loss factor. It was reported that the interface region hadprofound influence on deciding the effective dielectric properties of the composite systems [7]–[9].

The increasing addition of filler in the composite turn the composite system increases leading to poor packing and formation of porosity occur. This resulted in the existence of increased airspace and led to more air and water in the composite[7].

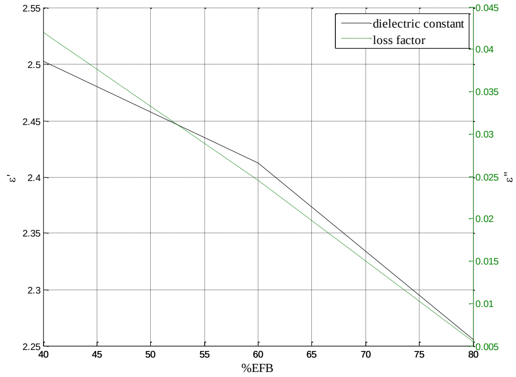

Fig.7a. Variation of dielectric constant and loss factor as a function of filler loading at frequency 8GHz

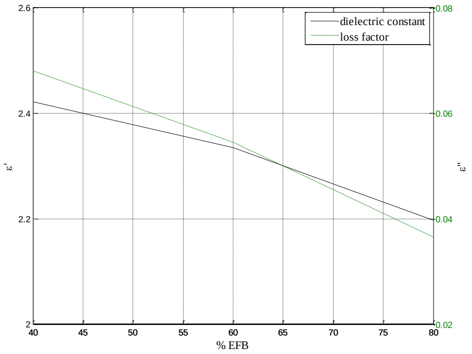

Fig.7b. Variation of dielectric constant and loss factor as a function of filler loading at frequency 9GHz

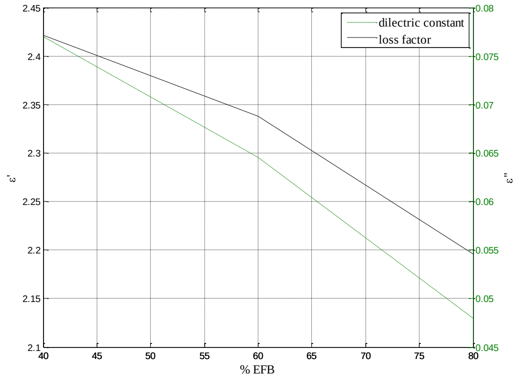

Fig.7c. Variation of dielectric constant and loss factor as a function of filler loading at frequency 10 GHz

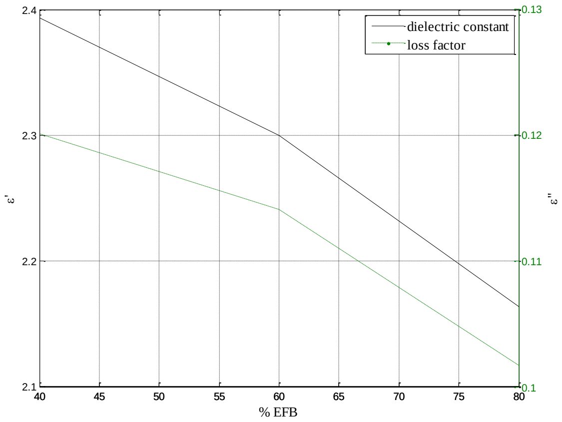

Fig.7d. Variation of dielectric constant and loss factor as a function of filler loading at frequency 11 GHz

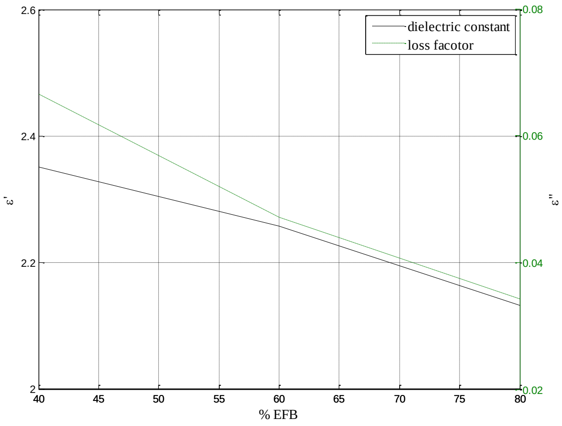

Fig.7e. Variation of dielectric constant and loss factor as a function of filler loading at frequency 12 GHz

Dielectric mixture model is the most commonly used theoretical model to calculate the complex dielectric constant of heterogeneous materials. The dielectric mixture is describedin terms of the fractional volume and dielectric constant of each constituent. For a composite system consisting of a mixture heterogeneous, dielectric constant optimum value is between the DC and the DC filler matrix. Nowadays models that are widely used are the Maxwell-Wagner-Sillar [9],Kraszewski [11] and Licthtenecker [8,11]as in Equations (1)–(3). These models enable material scientists to reach at novel composite systems without much experimental iterations[8].

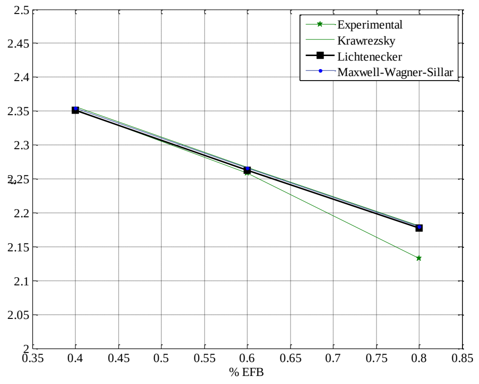

Fig.8: Comparison of theoretical predicted and experimental dielectric constant of composite

$$\log \varepsilon^* = V_f \log \varepsilon_f + V_m \log \varepsilon_m \qquad (1)$$

$$\varepsilon^* = \varepsilon_m \frac{2\varepsilon_m+\varepsilon_f+2V_f(\varepsilon_f-\varepsilon_m)}{2\varepsilon_m+\varepsilon_f-V_f(\varepsilon_f-\varepsilon_m)} \qquad (2)$$

$$\sqrt{\varepsilon^*} = V_f\sqrt{\varepsilon_f} + V_m\sqrt{\varepsilon_m} \qquad (3)$$

The notation used here applies to two components composite which are filler and matrix where ε^*represent the complex dielectric constant of composite, $\varepsilon_f$is the dielectric constant of filler and $\varepsilon_m$is the dielectric constant of matrix. The $V_f$and $V_m$are the volume ratio of the filler and matrix, respectively where$V_f + V_m = 1$ as the composite consist of two components. The theoretically predicted dielectric constant using these models was compared with that of experimental results. According to figure 6, all models almost matched the experimental data especially at lower filler loading. Comparing all the three models with the experimental results, it can be inferred that Kraszewski model has the best matching than other models. At the higher filler loading, all models have the same trend which deviates from the experimental results. This slight variation may be due to the porosity present in the samples.

IV. CONCLUSION

The PU-EFB composite was prepared with different volume of EFB filler through mixing and hot pressing. The crystallinity of the composite was finalized by powder X-ray diffraction studies. The microwave dielectric properties of the PU-EFB composites at X-band had been measured using vector network analyzer. Among different theoretical modelling studies attempted, Kraszewski model showed the best matching with experimental data.

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