Significance of Oxirane Rings in Epoxidized Palm Oil and Effects on the Coating Performance: Comparison between Epoxidized Unripe Palm Oil and Epoxidized Used Cooking Oil towards Adhesion Performance

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Abstract: *In this study, epoxy coatings made up from petroleum-based epoxy resin mixed with two different types of palm oils were prepared and compared. The commercial epoxy resin based on diglycidyl ether of bisphenol A (DGEBA) were formulated with epoxidized unripe palm oil (EPO) and another one with epoxidized used cooking oil (ECO) at four different ratios (0:100, 10:90, 20:80 and 30:70). The EPO and ECO were prepared through generated in situ of performic acid (HCOOH) and hydrogen peroxide (H$_2$O$_2$) by a fixed molar ratio of 1:5:4 mol/mol (EPO/ECO: HCOOH:H$_2$O$_2$). The reaction took place at constant temperature (45 °C) with continuous stirring for 150 min to obtain 95.5% oxirane conversion with 90.0% yield. The EPO and ECO were characterized by using Fourier Transform Infrared (FTIR) spectroscopy analysis. X-Cut and Cross-Cut methods were used in the test performance of adhesion strength. The results obtained from adhesion test for EPO were good as the coatings only experienced small trace of peeling. On the other hand, only minimal performances were obtained from the ECO coated metal. This was due to the presence of oxirane in EPO as compared to that of in ECO. EPO has a potential to replace petroleum-based resin in epoxy coating formulation according to its remarkable performances.*

Keywords: *epoxidized unripe palm oil; epoxidized used cooking oil; epoxy coating; epoxidation; adhesion performance*

■ INTRODUCTION

Bio-based materials recently received more attentions in product synthesis and manufacture especially in paint and coating industry. Escalated concern on depletion of fossil fuel and petroleum price has sparked widespread interest in alternative energy. Furthermore, the harm caused to the environment due to excessive use of fossil fuel has also contributes into awareness for more sustainable and environmentally friendly alternatives. Due to this, a research has been made by introducing bio-based materials into epoxy coating formulation in order to minimize or eliminate the dependency towards petroleum-based materials. However, bio-based materials have lower properties than the petroleum-based materials. Thus, in order to sustain the level of mechanical properties, the coating formulation was formulated by varying the ratio between the bio-based materials and petroleum-based materials.

Epoxy resin is known as the best barrier-coating material in industry due to their toughness, flexibility and high resistance to chemical [1]. Diglycidyl ether of bisphenol A (DGEBA) is a common epoxy resin used in formulating epoxy coating due to their high strength, excellent corrosion protection, and good adhesion to most surfaces [2]. However, DGEBA is carcinogenic that can cause tumor [3] and could affect men reproductive system [4]. The structure of DGEBA is shown in Fig. 1.

Moreover, DGEBA is a petroleum-based product which is poorly biodegradable and usually causing increased in carbon dioxide emissions during the manufacturing process. Therefore, consumers are now turning to eco-friendly ingredient as the main component
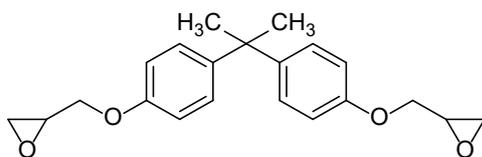
Fig 1. Structure of DGEBA [5]

in their coating material for better environment and health. Bio-based materials such as vegetable oils are safer and more beneficial because they are renewable, easily biodegrade and does not release a significant amount of greenhouse gas during manufacturing process [6].

The past history shows that the utilization of polymers based on vegetable oils such as *Annona squamosa* oil [7], *Mesua ferrea* L. seed oil [8], soybean oil [9], and palm oil [10] drew a great amount of attentions and supports from researchers, technologist, manufacturers and users. Vegetable oils are an excellent choice for the synthesis of bio-based polymers as they are biodegradable, non-toxic, non-depletable, non-volatile and most importantly low-priced and readily available in large quantities. As one of the world's leading producer of palm oil, Malaysia has high availability of palm oil. Palm oil is a type of edible vegetable oil that is produced from palm fruit of oil palm tree. Palm oil mainly consist of triglycerides, vitamin E, carotenoids, phytosterols, phospholipids, free fatty acids, gums, and oxidation products [11-12].

To increase the acceptability of palm oil for human consumption, palm oil undergoes refining process to remove the impurities such as undesirable odor, flavor, and color. The refined palm oil is so-called palm olein. Palm olein is largely use as cooking oil because of its stability at high temperature while at the same time has high antioxidant and limited amounts of unsaturated fatty acids [12]. Palm olein consists of oleic acid (39–45%) and linoleic acid (10–13%). Unsaturated fatty acids and saturated fatty acids also exist in approximately equal proportion in palm olein [13]. Repetition in using cooking oil during cooking and frying can cause deterioration of cooking oil making it no longer usable [14]. Cooking oil eventually is thrown away as waste without proper treatment. The accumulation of disposed used cooking oil polluted the environment and caused

problem to sewage system because at low temperature, the oil will be solidified and clogged the sewage system [15].

In oil palm productions, quality is always be determined by its texture, shape and color. In general, palm fruits turn from dark purple to reddish orange at the apex of their ripening. The rule of thumb for good quality practices is that, only ripe fresh fruit bunch (FFB) of oil palm are harvested and then dispatched to mills within 24 h after harvesting. The problem of poor quality FFB in the market can be minimized if they abide by the rules. However, in the FFB market, this basic principle is not strictly followed. Producers fail to ensure that only ripe FFB are harvested and left them mixed with the unripe FFB. The usual oil extraction rate for a ripe fruits bunches from a mature tree is between 22–24 percent, or 220–240 kg of oil per tonne of fresh fruit bunches [16]. On the contrary, under-ripe fruits bunches contain less oil due to long stalks that soaked up the oil and therefore reduce the oil extraction rate. In order to increase the oil recovery in palm oil mill, the extraction of residual oil from unripe FFB is important. Based on these facts, in this work the used cooking oil and unripe fresh fruit bunch of palm oil is used to develop a new coating material through epoxidation process and subsequently evaluate the resulting coating film's properties and performances.

Epoxidation process is a process where the unsaturated carbon bond present in the palm olein is chemically modified into a value-added product by addition of oxirane ring (Fig. 2). It is desirable to achieve a high oxirane oxygen value and lower iodine value as these criteria are considered to be of better quality [17]. The epoxidation process involves electrophilic addition mechanism as shown in Fig. 3. Epoxidation reaction as
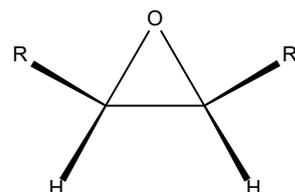
Fig 2. Structure of oxirane ring [10]
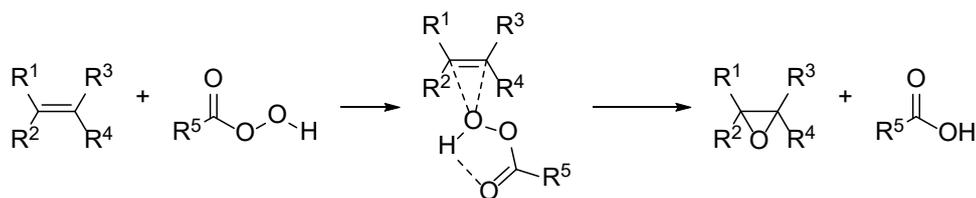
Fig 3. Epoxidation of unsaturated bond [17]
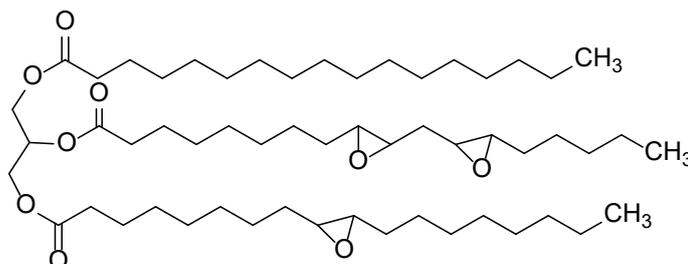
Fig 4. Structure of triglyceride after epoxidation with oxirane highlighted in green
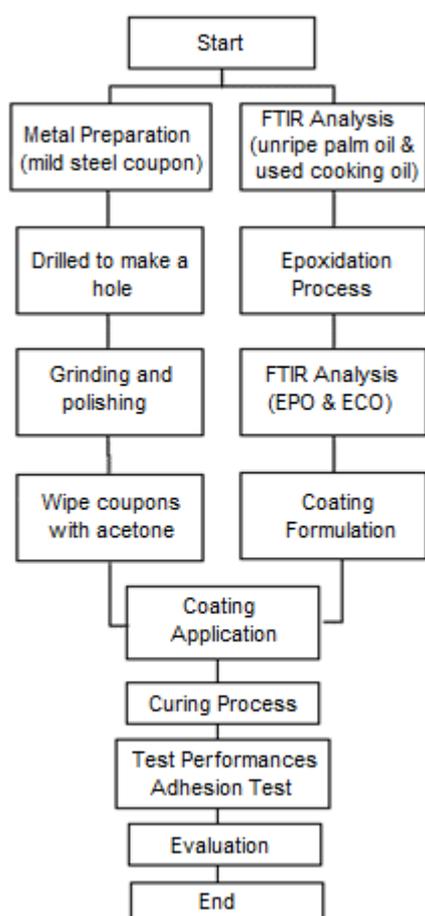
Fig 5. Overall process flow

shown in Fig. 3 takes place in similar way with unsaturated fatty acid structure. Oxirane ring forms at the unsaturated bond (C=C) to produce an epoxidized palm olein as shown in Fig. 4.

In this research, the epoxidized unripe palm oil (EPO) and epoxidized used cooking oil (ECO) will act as an additional material to reduce the toxicity of the current epoxy coating formulation. The bio-based epoxy coating was produced by varying the formulation into four different ratios (10:90, 20:80, and 30:70) of EPO/ECO to DGEBA. The overall goal is to develop a new coating material based on epoxidized unripe palm oil (EPO)/epoxidized used cooking oil (ECO) and Diglycidyl Ether of Bisphenol A (DGEBA) as well as to evaluate the performance of the new coating material based on their adhesion performance.

■ **EXPERIMENTAL SECTION**

Test panel for coating performance testing was prepared early before coating application. Unripe palm oil and used cooking oil undergoes epoxidation process before can be used in coating formulation. Observation was made to evaluate the performances for each coating with different formulation. Fig. 5 shows the general steps for this process.

Materials

Formic acid (98–100% purity) and titanium (IV) oxide powder were bought from Merck, Germany. Cycloaliphatic amine adducts (Isophorone diamine with

≥ 99% purity) and diglycidyl ether of bisphenol A were purchased from Sigma-Aldrich, USA. Used cooking oil was obtained from domestic waste. Unripe palm oil was obtained from Prof. Dr. Alawi Sulaiman from Faculty of Plantation and Agrotechnology UiTM Shah Alam. Hydrogen Peroxide (≥ 99% purity) was supplied by the chemistry laboratory in Faculty of Chemical Engineering, UiTM Shah Alam. Riken sandpaper grade 80, 180 and mild steel plate with dimension 60 × 60 × 3 mm were bought from hardware store in Seksyen 7, Shah Alam.

Instrumentation

Fourier Transform Infrared Spectrometer (Brand: Perkin Elmer Model: Spectrum 100), TQC Sheen wet film thickness gauge (Test Kit CC1000) follows ASTM D4414, ASTM D1212 & ISO 2808, DPM-816 digital coating thickness tester, TQC Sheen Cross-Cut cutter, mixer, dip-coater machine, acrylic container, knife, steel ruler and grinding/polishing machine.

Procedure

Epoxidation of unripe palm oil and used cooking oil

A molar ratio of unripe palm oil/used cooking oil:formic acid:hydrogen peroxide (1:5:4 mol/mol) was used in this epoxidation process with the mixture was heated and maintained at 45 °C [17]. Formic acid and hydrogen peroxide were mixed simultaneously in other container before added into the unripe palm oil/cooking oil. During addition of formic acid and hydrogen peroxide into unripe palm oil/used cooking oil, the addition action was done by slowly by adding drop wise of the mixture into the unripe palm oil/used cooking oil. The mixture was mixed homogeneously by using mechanical mixer for 150 min. A sample of unripe palm oil and used cooking oil before and after epoxidized were taken for Fourier Transform Infrared Spectroscopy (FTIR) analysis. The spectrum of unripe palm oil and used cooking oil before and after epoxidized were then compared and analyze for the presence of oxirane ring.

Characterization of palm oils derivatives

Fatty acid profiles of oils were obtained by using a FTIR spectrometer (Perkin Elmer Model; Spectrum 100) equipped with a deuterated triglycine sulphate (DTGS) as a detector and a KBr/germanium as beam splitter, interfaced to Computer operating under Windows-based, and connected to software of the OMNIC operating system (Version 7.0 Thermo Nicolet). A few drops of each sample were positioned in contact with attenuated total reflectance (ATR) on a multi-bounce plate of crystal at controlled ambient temperature (25 °C). The ATR plate was carefully cleaned in situ by rubbing with acetone and dried with soft tissue before filling in with the next sample, and made it possible to dry the ATR plate. The plate cleanliness was verified by collecting a background spectrum and compared to the previous one. These spectra were recorded as absorbance values at each data point.

Formulation of coating

Blend of epoxidized unripe palm oil/used cooking oil and epoxy resin were prepared with a weight ratio 0:100, 10:90, 20:80, and 30:70 (EPO/ECO:DGEBA) in different beakers. The mixture was mixed using mechanical stirrer at 400 rpm until no phase separation was observed. Then, titanium dioxide powder which act as a pigment was added into all the binder mixture produced at weight ratio of 1:0.5 (Binder:TiO_2). The mixture was then mixed using mechanical mixer at mixer speed of 1010 revolution per minute (rpm) until homogenous phase obtained at approximately 1 h. Finally, cycloaliphatic amine adduct as a crosslinking agent was added at a weight ratio of 1:0.25 (binder:amine). The mixture of binder and amine was then mixed using mechanical mixer at 400 rpm until no phase separation was observed.

Preparation of test panel

Sand paper no. 80 was used to remove any layer of rust and pitting from the surface of test panel. Test panels were then lightly abraded using sandpaper no. 180 and wiped with acetone to remove metal dust that was produced from abrasion before coating application.

Coating application

Coating was applied using Motor-Driven Dip Coater RDC 15 Application as referred to ASTM D823-95. A custom-made acrylic container was used in order to effectively coat the test panel and in the same time without using excessive amount of coating material. The

coating speed was kept constant for all test panels to maintain uniform thickness among all coating. Wet film thickness was measured using TQC Sheen wet film thickness gauge. Three readings were taken at each surface of test panel. Once desired wet film thickness was obtained, the coated test panels were left in laboratory under room temperature and standard condition to let the coating undergoes curing process at ambient temperature for 7 days [10].

Coating performance test
Adhesion test. After the coated test panel cured for 7 days, adhesion performance test was done in order to determine the degree of adhesion. X-Cut and Cross-Cut Tape Test were used to rate adhesion performance by referring to ASTM D3359-09. The adhesion performance was rated based on classification provided in ASTM D3359-09 in Table 1 and Table 2.

■ RESULTS AND DISCUSSION

Characterization Tests

Fourier transform infrared spectroscopy (FTIR) technique was performed to identify the chemical bonds existed in the unripe palm oil and used cooking oil before and after epoxidation process.

Table 1. X-Cut Tape Test adhesion classification

Classification	Explanation
5A	No peeling or removal
4A	Trace peeling or removal along incisions or at their intersection
3A	Jagged removal along incisions up to 1.6 mm (1/16 in.) on either side
2A	Jagged removal along most of incisions up to 3.2 mm (1/8 in.) on either side
1A	Removal from most of the area of the X under the tape
0A	Removal beyond the area of the X

Table 2. Cross-Cut Tape Test adhesion classification

Classification	Percent area removed	Surface of Cross-Cut area from which flaking has occurred for six parallel cuts and adhesion range by percent
5B	0% None	
4B	Less than 5%	
3B	5 15%	
2B	15–35%	
1B	35–65%	
0B	Greater than 65%	
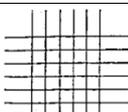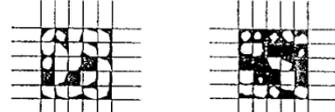
The spectrum of unripe palm oil as shown in Fig. 6(a) showed a stretching vibration peak of =CH (3003 cm⁻¹, a stretch of C–H was observed between 2921–2852 cm⁻¹, a carbonyl stretch C=O at 1743 cm⁻¹, and unsaturation peak HC=CH (cis) at wavenumber 1650 cm⁻¹. The peak at 3003 cm⁻¹ corresponds to the level of unsaturation that disappears as the double bonds are converted and used; thus, correlating with the epoxide peak at 844 cm⁻¹ as shown in Fig. 6(b). The same result generated was also demonstrated in the previous research [17]. The formation of epoxy group in the chemical structure of the epoxidized unripe palm oil indicates that the epoxidation process was successfully done.

Fig. 7(a) shows the spectrum profile of cooking oil before undergoes epoxidation process. A stretch of C–H was observed between 2921–2852 cm⁻¹. The spectrum shows carbonyl stretch at 1743 cm⁻¹ and methyl bending at 1464 cm⁻¹. Presence of nitrogenous compound is shown at 1542 cm⁻¹ and methylene group at 721 cm⁻¹. Bending of C-H was observed at 1377 cm⁻¹. Existence of C-O stretching and bending of O-H were found at 1159 cm⁻¹ and 1116.95 cm⁻¹ respectively.

While, Fig. 7(b) shows the spectrum profile of cooking oil after the epoxidation process. Based on the spectrum profile of ECO, there is no oxirane ring formed in the fatty acid structure from the epoxidation process as the oxirane ring should produce a trough between 833–841 cm⁻¹ [18-19]. Fig. 8 shows the summary of adsorption band of unripe palm oil and used cooking oil before and after epoxidation. Used cooking oil that was
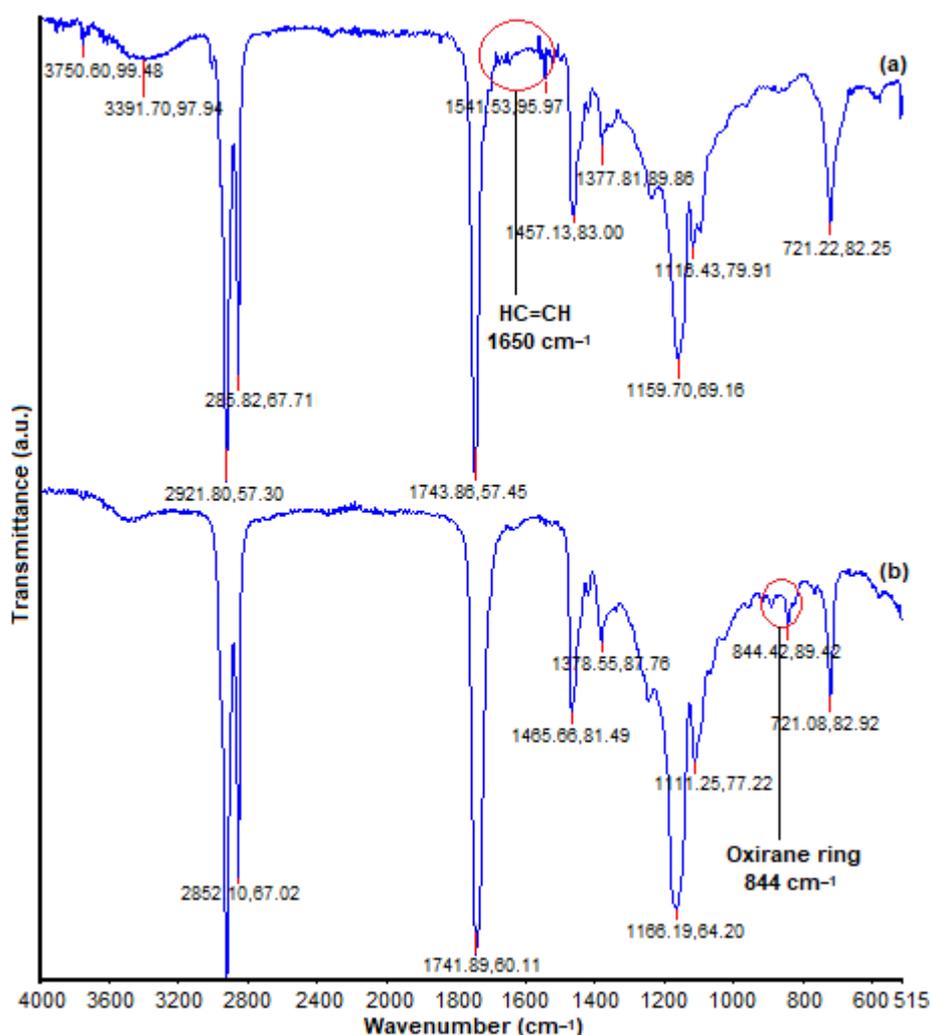
Fig 6. FTIR spectrum of (a) unripe palm oil and (b) epoxidized unripe palm oil
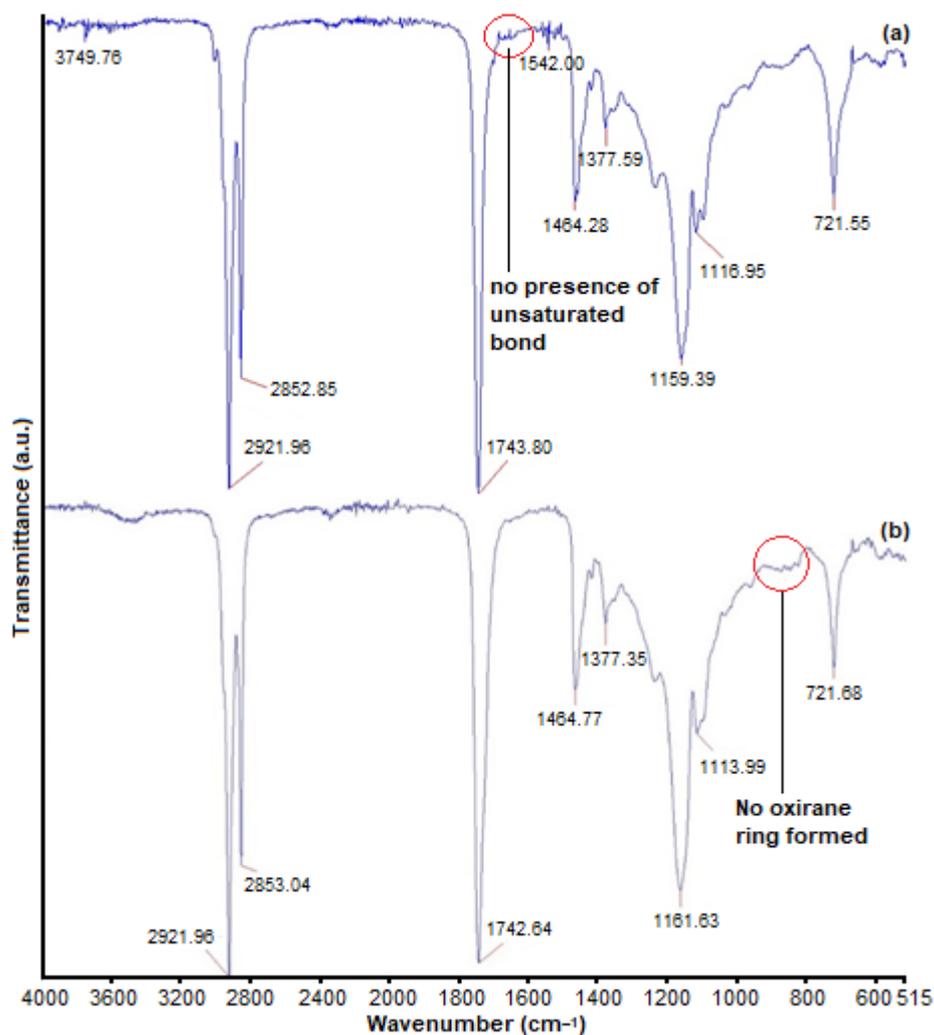
Fig 7. Spectral analysis of (a) used cooking oil and (b) epoxidized used cooking oil

Table 3. Summary of absorption band of unripe palm oil and used cooking oil before and after epoxidation process

Group	Unripe palm oil (cm^{-1})	Epoxidized unripe palm oil (cm^{-1})	Used cooking oil (cm^{-1})	Epoxidized used cooking oil (cm^{-1})
N-H str	3391	3394	3749	-
=CH str	3003	-	2921	2921
HC=CH (cis)	1650	-	-	-
C=O	1743	1741	1743	1742
Oxirane ring	-	844	-	-

collected from domestic household might consist of very low amount of unsaturated fatty acid or none at all. Existence of unsaturated fatty acid in the cooking oil is crucial for epoxidation process because unsaturated fatty acid provide reactive site for epoxidation process to complete [20]. Non-existence of unsaturated fatty acid in

cooking oil is due to decomposition process of unsaturated fatty acid during frying process [14-15].

Adhesion Performance Test

The coated test panel undergoes adhesion performance test in order to determine the degree of

adhesion performance. X-Cut and Cross-Cut Tape Test were used to rate adhesion performance by referring to ASTM D3359-07 [21]. The adhesion performance was rated based on classification provided in Table 1 and 2 as shown earlier.

X-Cut Tape Test

The results obtained from the X-Cut Tape Test for epoxidized unripe palm oil (EPO) coated metals were good as the coatings only experienced small trace of peeling as seen in Table 4. Formulation 0:100 serves as control which produced best performance with adhesion classification as 5A. Formulation 10:90 and 20:80 are both classified as 4A. Formulation 30:70 also produced the best adhesion classification which is 5A. On the other hand, results from X-Cut Tape Test for ECO coated metals showed that only minimal adhesion performances obtained from ECO coated metal for all formulation. If observed and compared among the coating that was added with ECO in their formulation, 10:90, 20:80 and 30:70,

Table 4. Results of adhesion X-Cut Tape Test of coated metal of EPO and ECO

Ratio	EPO:DGEBA	Classification	ECO:DGEBA	Classification
0:100		5A No peeling or removal		5A No peeling or removal
10:90		4A Trace peeling or removal along incisions or at their intersection		1A Removal from most of the area of the X under the tape
20:80		4A Trace peeling or removal along incisions or at their intersection		0A Removal beyond the area of the X
30:70		5A No peeling or removal		2A Jagged removal along most of incisions up to 3.2 mm (1/8 in.) on either side
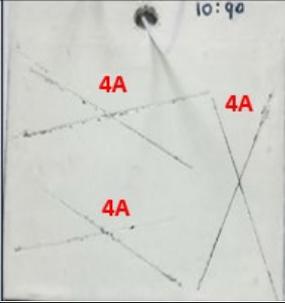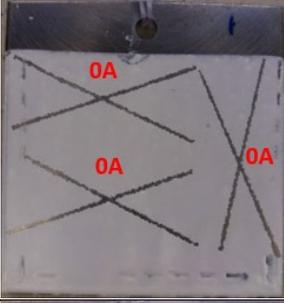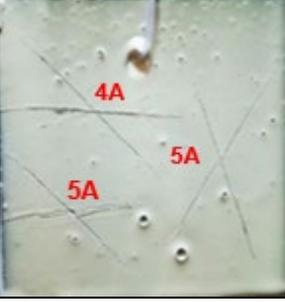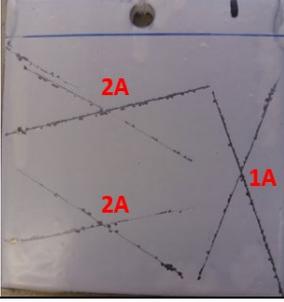
they obtained almost similar adhesion classification. Only formulation 20:80 yielded one rank lower than 10:90 and 30:70. There is no distinctive trend can be observed in result obtained from X-Cut Tape Test.

Cross-Cut Tape Test

Table 5 shows the condition of coated metal of EPO and ECO after being tested with Cross-Cut Tape Test.

The average percent area removed of coated metal was observed and calculated, then classified according to the standard ASTM D3359-09 in Table 2. As can be seen in Fig. 8 and Table 6, formulation 10:90(EPO: DGEBA) was classified as 3B with minimum error bar of 2B, formulation 20:80 was classified as 4B with minimum error bar of 3B and formulation 30:70 was classified as 4B with maximum error bar of 5B. Addition of EPO in the

Table 5. Result of adhesion Cross-Cut Tape Test of coated metal of EPO and ECO

Ratio	EPO:DGEBA	ECO:DGEBA
0:100		
10:90		
20:80		
30:70		
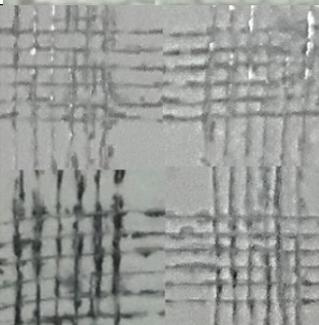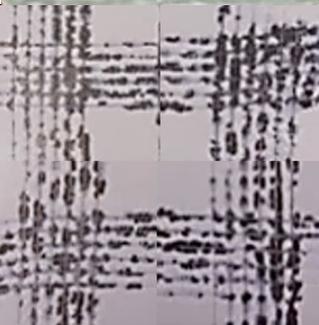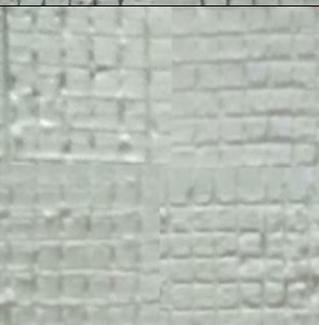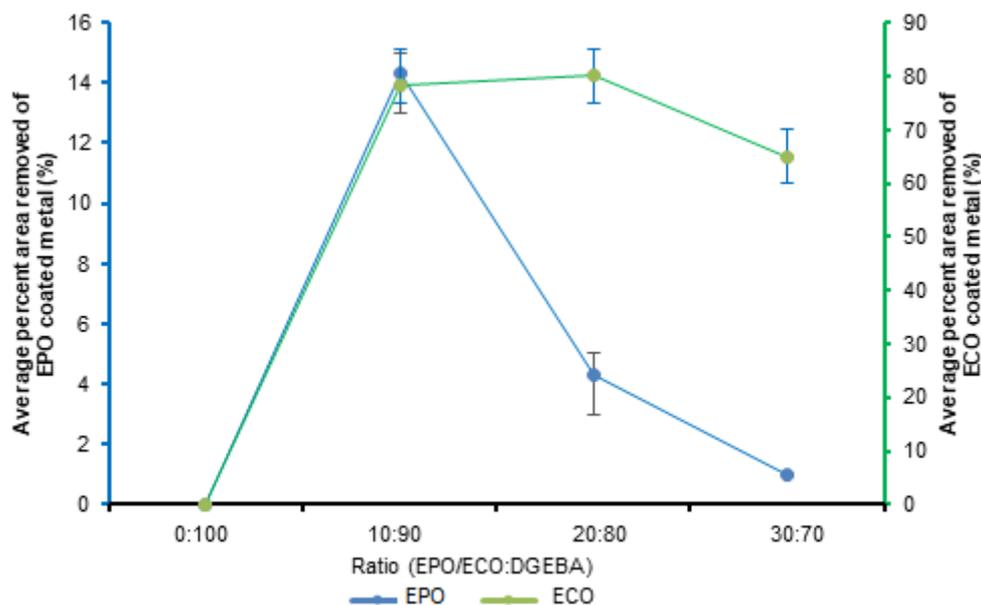
Fig 8. Graph of adhesion performance of developed coating by Cross-Cut Tape Test

Table 6. Summary of Cross-Cut Tape Test adhesion classification

EPO:DGEBA		
Ratio	Average Percent Area Removed (%)	Classification
0:100	0	5B
10:90	14.33	3B
20:80	4.33	4B
30:70	1.0	4B
ECO:DGEBA		
0:100	0	5B
10:90	78.33	0B
20:80	80.0	0B
30:70	65.0	1B

coating formulation have reduced the brittleness of the coating, as the components of EPO in the coating formulation increase, the adhesion performance become better. Meanwhile, only minimal adhesion performance obtained for all formulations of ECO:DGEBA. Both formulations (10:90 and 20:80) which was tested by Cross-Cut tape test were classified as 0B and formulation 30:70 was classified as 1B. These adhesion results support the findings from FTIR analysis of epoxidized cooking oil in Fig. 7(b) that there is non-existence of oxirane ring in the epoxidized cooking oil which is important component to develop a good coating material.

■ **CONCLUSION**

In this study, the unripe fruit bunch and used cooking oil was successfully utilized as additional compound in new epoxy formulation. Incorporation of these oils as additional component in formulating new epoxy coating gives oil palm a new area of application and adds value to this renewable resource. Comparison of FTIR analysis on unripe palm oil and used cooking oil before and after epoxidation show different results. Oxirane ring was present at a wavenumber 844 cm^{-1} in EPO whereas the oxirane ring was absent in ECO. The results obtained from the adhesion test from EPO were

good as the coatings only experienced small trace of peeling. Only minimal adhesion performances obtained from ECO coated metal for all formulation. This correlated with absence of oxirane ring from the ECO observed in spectrum profile. Finally, it may be concluded that epoxidized unripe palm oil has a potential to replace petroleum-based resin in coating formulation based on their good performances.

■ ACKNOWLEDGMENTS

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