



Optimization of Curing Process for Production of Jatropha Oil Bio-Based Resin Decorated With Nano- Al_2O_3 and Mechanical Characterization

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HIGHLIGHTS

- The production of green and renewable bioresin from jatropha oil decorated with Aluminium oxide nanoparticles
- Epoxidized crude jatropha oil (ECJO) and nano- Al_2O_3 particles are being used to optimize the property of a cured ECJO based resin consisting of 25% ECJO and 75% synthetic polymer.
- A range of 0% to 4% of nanoparticles was tested on epoxy bio-resin. Specimens fabricated were characterized for its mechanical properties.
- Addition of nano- Al_2O_3 improved tensile strength of a ECJO resin with its optimum at 1 wt% addition reaching a tensile stress of 29.37 ± 2.00 MPa and elastic modulus of 840.80 ± 124.53 MPa.

ABSTRACT

Concerns on the environment, health and safety issues posed by synthetic resins has amplified numerous efforts of producing resin from various renewable sources. The use of plant oil as potential source of resin has attracted interest from various researchers. Jatropha Oil is a competitive source to petroleum counterparts due to its availability, biodegradability, low eco-toxicity but exhibit poor mechanical properties among many. The objective of this study is to study the effect of adding nanoparticles as reinforcing fillers to bio-based resin from Epoxidized crude Jatropha Oil (ECJO) to improve its mechanical performance. Various loadings of 0% to 4% of Al_2O_3 nanoparticles was tested on epoxy bio-resin. Later the specimens fabricated were cured and characterized for its mechanical properties. Addition of 1 wt% of Al_2O_3 nanoparticles improved the tensile strength of a bio-based epoxy resin to tensile stress of 29.37 ± 2.00 MPa, elastic with an elastic modulus of 840.80 ± 124.53 MPa. Further characterization at optimum addition of nano- Al_2O_3 resulted a glass transition temperature of 37.95°C . In overall, the inclusion of nano- Al_2O_3 has definitely improved the mechanical properties of the material which will be useful for further application material engineering.

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1. Introduction

‘Epoxy resin’ is a term used for the prepolymers and its cured resins in which the former contain reactive epoxy groups Gilbert [1]. In cured resins, cross-linking occurs at epoxide group of polymer chain by reacting with themselves or co-reactants (curing agents) comprising of amines, anhydrides, phenols, and thiols among many others in order to produce a thermosetting polymer with high mechanical properties, temperature resistance and chemical resistance. Demand of epoxy resins is driven by the growth in end user industries such as paints and coatings, electrical and electronics, construction, composites and adhesive industries.

Sustainable development has become a large focus of research globally. Due to environmental concerns and the depleting oil reserves research and efforts on biodegradable products and renewable resource has been widespread Dinda et al [2]. Growing interest in the use of materials from renewable origin has been detected as a result of this growing concern

on environment through recycling, attention to biodegradability or upgrading of materials. This is highlighted in the polymer composite industry where great amounts of petroleum-based polymers are used Bertomeu et al [3].

Polymers derived from renewable sources such as carbohydrates, starch, proteins, fats and oils have gained an increasing interest because of their low cost and biodegradability Soldo et al [4], Jin et al [5]. Plant oils such as palm oil, soybean oil, Jatropha oil, have received considerable attention as one of the renewable resources for not only the production of energy, but chemicals in general including starting materials for polymers. The epoxy resin industry is no exception as its source is still very much dependent on fossil resources and the search and important efforts are being done in the field high bio-based content resins

The glass transition temperature (T_g) is one of the most important product properties of an epoxy resin as it determines the window of applications. High T_g (100-150 °C) resins are typically required for structural adhesives and sheet molding compounds whereas low T_g ones (<30 °C) are suitable as adhesives for electronic and automotive applications.

Although the use of bio-resin can reduce environment pollution as it is completely degradable in nature, it often possesses drawbacks such as poorer mechanical properties, poorer processability and higher hydrophilicity that would limit their application Kumar, [6]. Bio-based materials had a rapid development and an increasing variety of resources such as vegetable oils, starch, soy protein and cellulose have been used to produce polymeric materials that have comparable quality in their properties to their petroleum-based counterparts. These bio-based materials such as polylactic acid (PLA), poly(hydroxyalkanoates) (PHA) and castor oil based polyamide have even been commercialized with their additional environmental value alongside that of petroleum-based. However, compared to the progress in bio-based thermoplastics, the research on bio-based thermosetting resins that possesses a comparable mechanical properties with petroleum-based counterparts are still limited Liu et al [7]. The properties of bio-resin can be enhanced by modifying the resin with the use of additives or fillers Kumar [6].

In the current study, an epoxy bio-resin from Jatropha Oil is produced and its cured blend is optimized by the addition of aluminum oxide nanoparticles (nano- Al_2O_3). To characterize the cured blend of epoxy resin, tensile test was performed to determine its mechanical property and glass transition temperature.

2. Materials and Method

2.1 Materials

Crude Jatropha Oil (CJO) from Bionas Sdn. Bhd., Amberlite IR-120, Glacial Acetic Acid, Hydrogen Peroxide (30%), anhydrous sodium sulphate, commercially available epoxy resin (EpoXAmite 100 Epoxy Laminating System) and Hardener (EpoXAmite 103 slow hardener) from Smooth-On, Inc., Aluminum Oxide Nanopowder (Al_2O_3 , alpha, 99+% 80nm) from US Nanomaterials, Acetone.

2.2 Production of Epoxidized Crude Jatropha Oil (ECJO)

In-situ epoxidation of crude Jatropha oil with aqueous hydrogen peroxide and acetic acid in presence of Amberlite IR-120 acidic ion exchange resin as catalyst was conducted. The epoxidation reaction took place in a 3 neck round bottom flask with an analog overhead stirrer with Teflon blade and a water bath temperature controller. A calculated amount of CJO (263 g, 1 mol), glacial acetic acid (39 g) and Amberlite IR-120 (42 g) were added to 1L three neck round bottom flask. Mixture in flask was heated up to 65 °C, which was 10 °C below the desired reaction temperature. 30 % H_2O_2 (194mL) was introduced to separating funnel. After temperature was achieved, addition of H_2O_2 was carried out at this temperature as reaction exothermic to prevent any hazard from excessive release of heat. Addition of H_2O_2 was done drop by drop for the same reason and agitation by overhead analog stirrer was performed using a Teflon impeller. Continuous stirring throughout reaction was maintained so as to avoid the formation of high concentration zones. After the complete addition of H_2O_2 , the temperature was increased to 75 °C and reaction was left for a total of 5 hours. The completion of H_2O_2 addition marked the zero hour. After 5 hours, the resulting mixture was transferred into a separating funnel draining any aqueous layer or catalyst. The product was washed with distilled water for three times at three different temperatures of distilled water; hot (~90°C), cold (chilled) and hot (~90 °C) to ensure the product was acid free. The ECJO was then further dried with anhydrous sodium sulphate in the ratio of 1:0.15 and heated at 80 °C for 12h. The drying process was to ensure the removal of trace water. The resulting mixture was filtered and any aqueous layer was disposed.

2.3 Curing of Epoxidized Crude Jatropha Oil (ECJO) using Nano- Al_2O_3

The curing and nano-dispersion method (solvent-based technique) has been adapted and adopted from Farah et al. [8], and Haq et al [9].

A composition of 75% of epoxy resin and 25% of ECJO and 100% epoxy resin for control was prepared. The amount of hardener needed to cure was in accordance to the supplier recommendation. Initially the nano- Al_2O_3 (0, 1, 2, 3, 4 wt%) were dispersed into 750 ml acetone to achieve as much as possible separated nanoparticles from each other by sonicating for a total of 15 minutes (Qsonica Sonicators). At the end of 15 minutes, this solution was mixed into EpoxAmite 100/ECJO blend for a total of 5 minutes. Mixture was then sonicated again using ultrasonic cleaner baths (WiseClean). Acetone was removed from the epoxy mixture by heating up mixture at 60°C.

Mixture was cooled down before addition of hardener because of exothermic mixing. The EpoxAmite 103 slow hardener was added accordingly to EpoxAmite 100/ECJO/Nano- Al_2O_3 . Resulting mixture was poured into a waxed surface using mold release agent and aluminum pan for molding. The thickness of the sample was to be maintained evenly at a range of 5 mm to 8 mm. At an even surface, sample was left for 20~24 hours to cure at room temperature. As a post-curing treatment, samples were cured for 2 hr at 60°C, 3 hr at 80°C, and 3 hr at 100°C.

2.4 Characterization of Nano- Al_2O_3 Incorporated Epoxy Resin/ECJO Cured Blend

After a successful curing, characterization of the cured blend involves the evaluation of its physical property by tensile properties. As a requirement for tensile testing, cured blend of bio-synthetic resins incorporated with Al_2O_3 nanoparticle was cut into standard sizes in accordance to ASTM D638 using a band saw machine. Dimensions of sample follows that of thickness of 5mm to 8mm, width of 250mm and length of 220mm. Tensile testing was performed with a speed of 4mm/min.

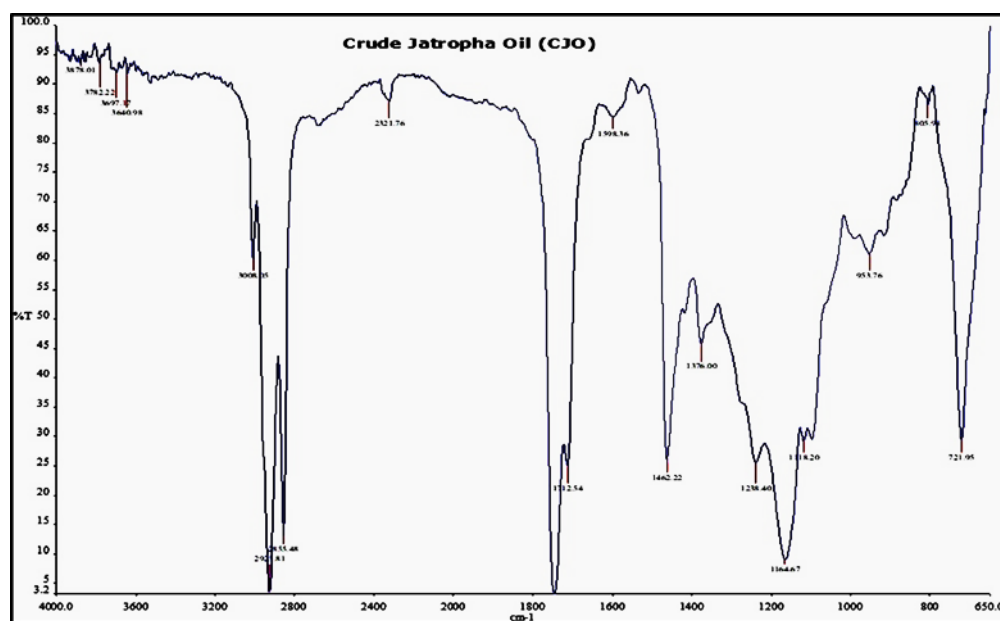
Differential scanning calorimetry (DSC) analysis was performed using a DSC823 from Mettler Toledo with a heating rate of 10 °C/min and cooling rate of 10 °C/min. The T_g was determined from the inflection point of the second scanning curve.

3. Results and Discussion

3.1 Fourier Transform Infrared Spectroscopy (FTIR) of Epoxidized crude Jatropha oil (ECJO) and Crude Jatropha Oil (CJO)

Epoxidized Crude Jatropha Oil was obtained from the epoxidation of Crude Jatropha oil acquired from Bionas Sdn. Bhd. Factors affecting the epoxidation reaction including temperature of reaction, amount of acetic acid, amount of hydrogen peroxide and the amount of the catalyst loaded were controlled throughout the production of ECJO according to determined condition

FTIR results allow ones to identify the functional group in any samples. Here the FTIR for CJO and ECJO after epoxidation process were monitored. Figure 1a illustrates the peaks for the unsaturated alkene (C-H) at 3008 cm^{-1} in the CJO compound. The disappearance of the double bond in the ECJO compound from the spectra and appearance of new peaks at 822 cm^{-1} confirmed the synthesis of the epoxides. This result was in confirmation with previous works that reported the band of oxirane formed in the range of 820 to 843 cm^{-1} Abdullah et al. [10], Ikhuoria et al. [11], Farah et al [8]. However, it is apparent from Figure 1b that the peak of the oxirane ring (C-O-C) was not very significant due to low epoxy contents in ECJO which might be due to only 60.55% of double bonds conversion to oxirane ring Farah et al [8]. The non-existence of the hydroxyl peaks (3000-3500 cm^{-1}) in the ECJO compound proved that the minimum oxirane decomposition occurred in the epoxidation reaction Farah et al [8].



(A)

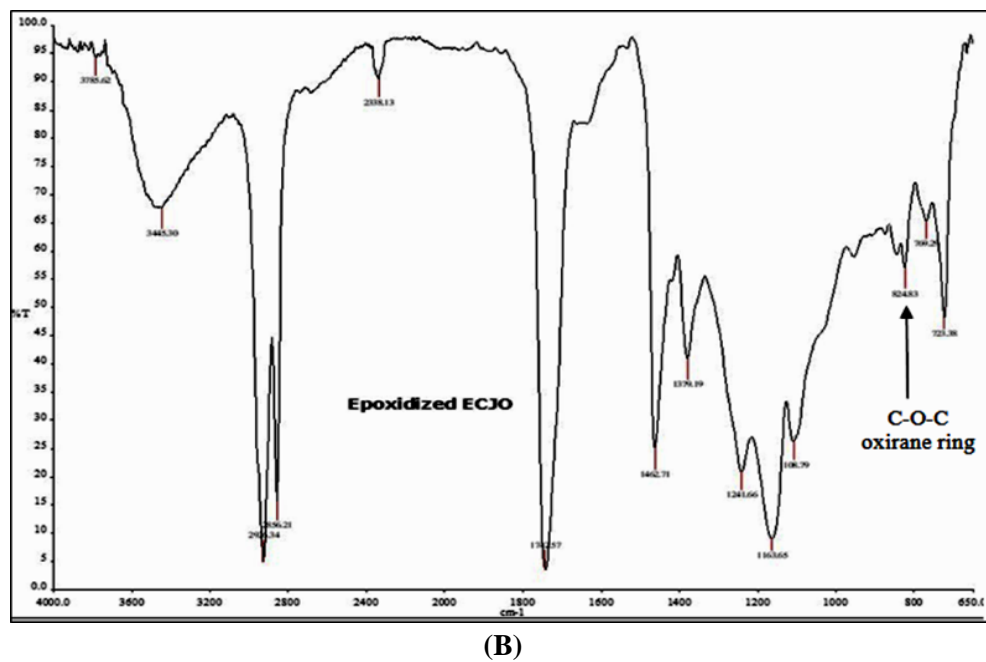


Figure 1: Illustrations of FTIR Spectra for (a) CJO and (b) ECJO

3.2 Mechanical Properties

3.2.1 Effect of Al_2O_3 nanoparticles and blending ECJO/synthetic resin on tensile strength

To investigate its tensile strength, a set of 5 specimens was prepared for epoxy and ECJO blend. 25:75 ratio of ECJO to synthetic epoxy resin was added with various loading of Al_2O_3 nanoparticles ranging from 0 - 4 wt%. In addition, for comparison, another 5 specimens of 100%:0% ratio of synthetic epoxy resin to ECJO with different loadings of Al_2O_3 nanoparticles was also prepared and studied.

The ultimate tensile stress of a material is also known as the tensile stress at break in which it is the maximum stress the material can withstand until fracture. The result trend for the tensile stress is depicted in Figure 2. Both results using 0% ECJO and 25% ECJO showed similar trends of behaviour whereby the tensile stress increased as the nanoparticles amount was increased to 1% and then decreases as the nanoparticle loadings reduced to 4%. At 0% Al_2O_3 content, the tensile stress for 0% ECJO and 25% ECJO was $41.42 \pm 6.55 \text{ MPa}$ and $10.08 \pm 0.14 \text{ MPa}$. The tensile stress trend showed that the optimum tensile stress for both 0% ECJO and 25% ECJO epoxies are at 1% Al_2O_3 nanoparticles addition. While the optimum tensile stress for 0% ECJO neat epoxy resin reaches up to $52.41 \pm 8.465 \text{ MPa}$ at 1% nano- Al_2O_3 , the optimum tensile strength achieved by 25% ECJO with 1% nano- Al_2O_3 is $29.37 \pm 2.00 \text{ MPa}$.

The increase of the mechanical performance for 25% ECJO is very drastic from EpoxAmite/ECJO blend without the addition of nanoparticles. After an increase of tensile strength at 1% nano- Al_2O_3 , adding more nanoparticles resulted in the decreasing of the mechanical performance to $29.37 \pm 2.35 \text{ MPa}$ and $8.90 \pm 0.77 \text{ MPa}$ at 2% nano- Al_2O_3 addition for 0% ECJO and 25% ECJO respectively which is slightly below the unmodified sample. Increasing nanoparticles content to 5%, cause an even greater drop of mechanical performance to $7.773 \pm 0.646 \text{ MPa}$ and $3.78 \pm 0.591 \text{ MPa}$ for 0% ECJO and 25% ECJO respectively.

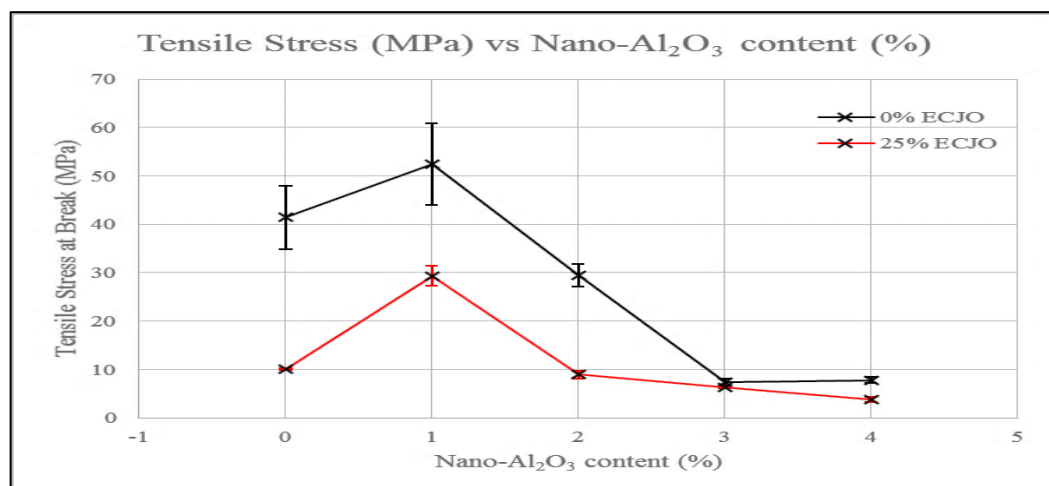


Figure 2: Tensile stress of cured EpoxAmite/ECJO/nano- Al_2O_3 resin

While the addition of nanoparticles can greatly enhance the performance of a polymer, it is shown that the higher the nanoparticles content does not necessarily lead to a higher improved mechanical property such as shown by our results. These findings conformed to several studies including by Omrani and Rostami [12], Zheng et al [13], and Wang et al [14].

The addition of Al_2O_3 nanoparticles lead to decrease in space distance between epoxy chains. Polar nanoparticles filling free space between chains and attracting resin molecules leads to more complicated network chains during curing process of epoxy resin such as creating hydrogen bonds between chains and particles. As a result, there is an increase of constraint between; particles/polymer chains, and polymer chains itself. This alter the space distance and increase free volume space which leads to lesser ability of epoxy chains to bear greater forces Kadhim et al [15]. Moreover, the behavior of decreasing strength may also be caused by nanoparticles aggregation/agglomeration Zare et al [16]. It increases with addition of nanofiller content and reduction of nanofiller size which unfortunately reduces the effectiveness of nanoparticles in polymer matrix and lead to poor properties of samples.

Other than agglomeration, inferior performance can be also due to the presence of pores at the interface between filler particles and the matrix. The interfacing adhesion may be too weak to transfer the tensile stress or corner points of irregular shaped particulates which results in stress concentration in the matrix base Sravani et al [17].

3.2.2 Effect of Al_2O_3 nanoparticles and blending ECJO/synthetic resin on elastic modulus

Elastic modulus is essentially a measurement given to indicate the resistance of material to tensile force. Figure 3 shows that at 0% nano- Al_2O_3 content, the elastic modulus for 0% ECJO and 25% ECJO was $1184.44 \pm 502.54 \text{ MPa}$ and $319.9235.32 \text{ MPa}$ respectively.

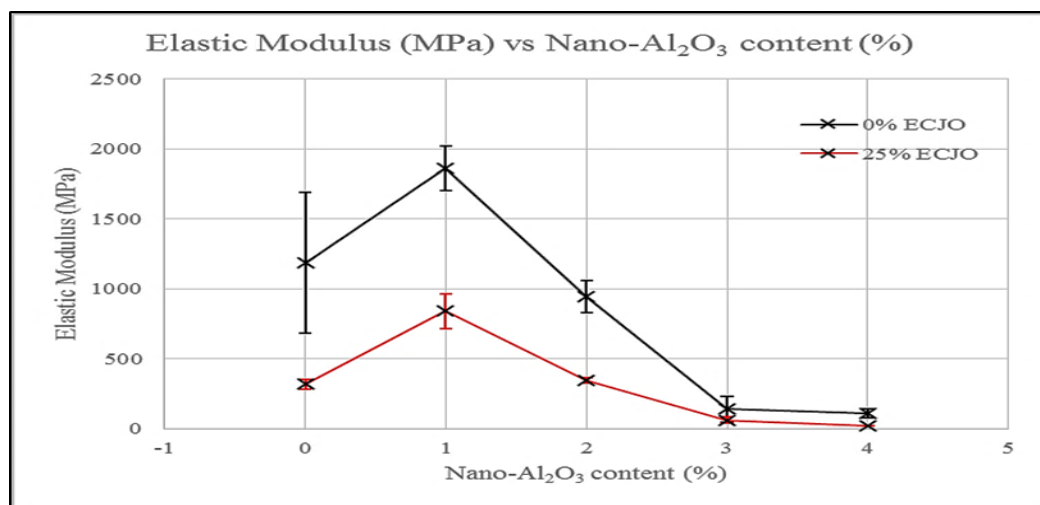


Figure 3: Elastic modulus of cured EpoxAmite/ECJO/nano- Al_2O_3 resin

Optimum condition is achieved at 1 wt% of nano- Al_2O_3 with an average modulus value of $1860.93 \pm 157.864 \text{ mPa}$ and $840.796 \pm 124.531 \text{ mPa}$ for 0% ECJO and 25% ECJO respectively. Similar to tensile stress behaviour previously, the elastic modulus displayed an increase with the addition of nanoparticles up to 1%wt and later decreases as the loadings increases to 4wt%. The specimen tends to be less strong, more elastic and susceptible to be elongated with tensile force. By theory, the increment in Young modulus is because nanoparticles can fill up the weak microregions of resins to boost the interaction forces Zheng et al [13].

Wang et al [14] also reported a rising trend of modulus with the addition of nanoparticle into epoxidized soybean oil. The same trend is also portrayed in study of mechanical properties of dental composite with nano- Al_2O_3 filler particles Foroutan et al [18]. However, Omrani [12]. found that the relationship between the flexural modulus with the addition of nanoalumina was remarkably increased when 0.5phr of the nanoalumina is added. The maximum flexural modulus was achieved when the addition of the filler was 2phr and later decrease at subsequent addition.

Cast epoxy resin usually has a tensile modulus ranging from 2400MPa-3500MPa, an ultimate tensile stress 30-90 MPa, and an ultimate tensile strain of 3-6%. As control study, we also investigated the stress of a neat epoxy blend which composed of synthetic epoxy and its hardener without the addition of ECJO or nano- Al_2O_3 (0%ECJO, 0% Al_2O_3). The results Figure 4 showed that this control samples possesses a tensile strength of $41.42 \pm 6.46 \text{ MPa}$, tensile strain of $6.80 \pm 2.67\%$, and a modulus of $1184.44 \pm 502.54 \text{ MPa}$. The action of adding of Al_2O_3 nanoparticles cause a drastic decline of mechanical properties to eventually record an ultimate tensile stress of 7 MPa, tensile strain of 27% and modulus 111 MPa at 4wt%.

3.2.3 Effect of Al_2O_3 nanoparticles and blending ECJO/synthetic resin on tensile stress and strain behaviour

Figure 4 depicts the tensile stress against tensile strain graph for the specimens of different conditions of nanoparticle content for epoxy resin without ECJO. Out of the 3-5 test specimens for every condition, one was chosen as a representative of the stress-strain relationship.

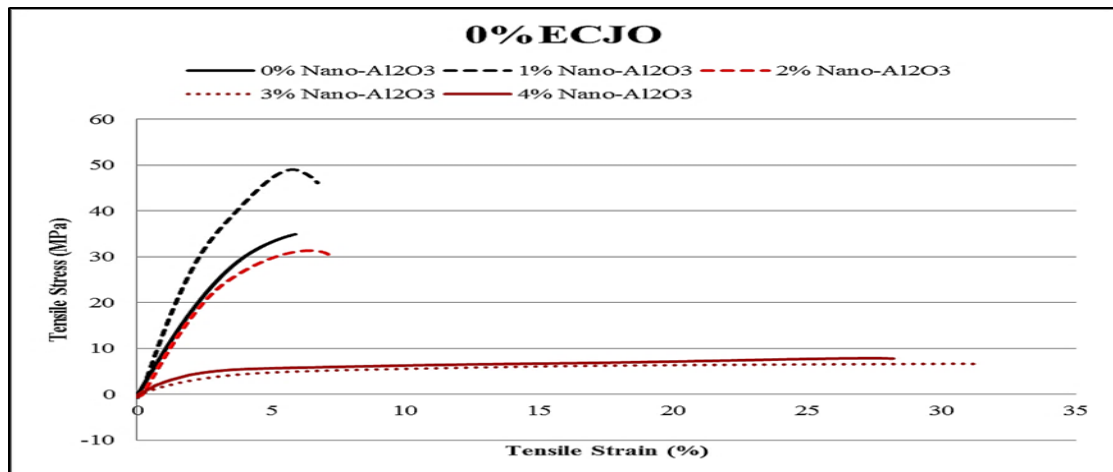


Figure 4: Stress-Strain graph for 0%ECJO at different nano-Al₂O₃ content

The neat epoxy resin without any nanoparticle follows the trend of that of glassy polymer. Glassy polymers are hard and relatively brittle. With the addition of 1 wt% and 2 wt%, it is observed that specimen still holds glassy properties with high ultimate tensile stress and ultimate tensile strain. The specimens with this trend are stronger in performance and is not easily susceptible to elongation. All three specimens (0 wt%, 1 wt%, 2 wt%) exhibit an ultimate tensile strain of $6.08 \pm 2.67\%$, $5.52 \pm 1.08\%$, and $6.44 \pm 0.14\%$ respectively. As shown in Figure 4, the 1 wt% addition gives a drastic improvement from stress and strain while the addition of 2 wt% nanoparticles drop below the mechanical properties of the unmodified epoxy resin.

After the addition of 2 wt%, the specimens tend to be less glassy in property. They are more disposed to elongate with a tensile force. The trends of both 3wt% and 4wt% addition of nano-Al₂O₃ exhibits more of a stress-strain diagram that represent ductile deformation behavior with higher strain of $31.59 \pm 1.96\%$ and $27.31 \pm 6.31\%$ respectively Moser & Feuchter [19]. The trend for both 3 and 4wt% composition resembles that of a tough material without yield point.

Previous studies suggested that blending the DGEBA based epoxy materials with aliphatic or cyclic epoxy-species results in a significant change in Young's modulus (elastic modulus), tensile strength and elongation. Bio-based alternative epoxies on the other hand exhibits a lower tensile strength and lower modulus Moser & Feuchter [19]. Hence, there is a limitation to which blending of synthetic to biobased epoxies can be done to minimize the impact it has on the mechanical properties. Figure 5 shows the stress-strain results of the 75:25 blending ratio of synthetic epoxy resin to ECJO with different loadings of Al₂O₃ nanoparticles.

Without the addition of nano-Al₂O₃, characteristic of the synthetic/ECJO blend shows a high tensile strain and tensile strength reaching up to $50.33 \pm 16.52\%$ and 10.08 ± 0.14 MPa respectively. On the other hand, a different behavior was observed upon the addition of Al₂O₃. At 1 wt% addition of Al₂O₃ nanoparticles, a sharp increase of tensile strength and decrease of tensile strain was achieved which is a typical trend of a stress-strain behavior corresponding to a glassy and brittle polymer.

Adding Al₂O₃ more than 2 wt% caused the tensile stress and strain to exhibit similar behavior as the unmodified specimen (0% nanoparticles). A greater nanoparticles loadings resulted in lower stress and strain values that resembled an elastomer or rubber. In short adding more nano-Al₂O₃ beyond 1wt% onwards causes decrease in tensile stress.

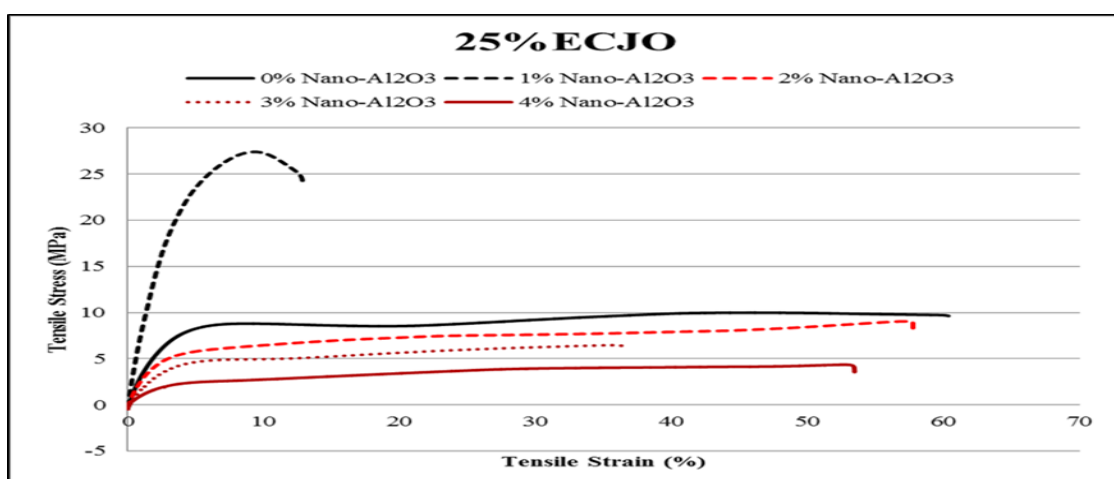


Figure 5: Stress-Strain graph for 25%ECJO at different nano-Al₂O₃ content

3.3 Glass Transition Temperature

In response to applied stress, polymeric materials including epoxy resins demonstrates not only an elastic property but also a viscoelastic property. Viscoelastic response is commonly known as viscoelasticity, and it plays an important role in the long-

term performance of an epoxy resin. The position of the viscoelastic transition region is a characteristic for determining the thermo-mechanical properties of these thermoset materials.

The change from the viscoelastic state to the glassy state and vice versa is defined as glass transition. The glass transition temperature of a polymer can be understood as a range of temperature where polymer molecules transition from a more organized, crystalline type of arrangement (usually at cooler temperatures) to the loss of rigidity, gradual softening to a more rubbery state (at higher temperatures). For the case of the optimum performing Al_2O_3 incorporated epoxy bio-resin according to tensile testing (25% ECJO, 1 wt% nano- Al_2O_3), the glass transition temperature of was recorded at 37.95°C Figure 6.

The commercially available epoxy resin when cured may go up to the normal range of glass transition temperature of 70 to 80°C. This has been reported by other researchers when the commercial epoxy resin containing carbon nano tube (CNT) and short carbon fibers was tested for its On the other hand, the action of blending DGEBA (commercial petroleum-based resin) with epoxidized soybean oil caused a significant reduction of Hence, previous findings conformed to our results. The addition of ECJO to commercial resin will decrease the glass transition temperature to a certain extent.

Curing schedule also plays another role in defining the final glass transition temperature. 0% of ECJO did not reach high T_g which indicates that other epoxy resin matrix maybe more suitable for this application. Lower T_g adhesives is more suitable for application such as in electronics.

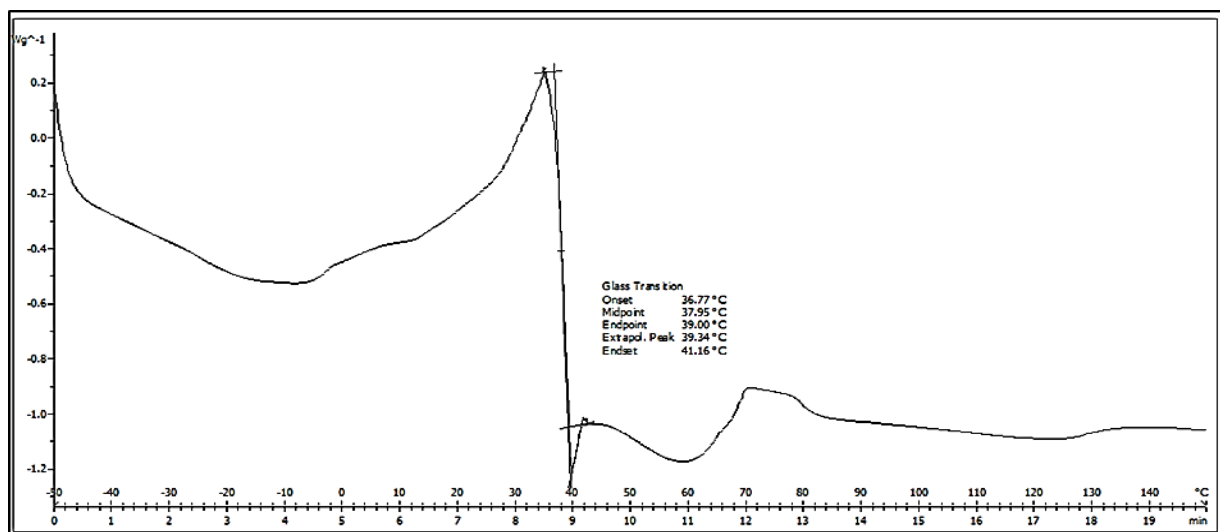


Figure 6: DSC curve for Epoxy/ECJO blend at 1 wt% nano- Al_2O_3

4. Conclusion

The epoxidized crude Jatropha oil (ECJO) was obtained from the epoxidation of crude Jatropha oil (CJO) by acid ion exchange resin (AIER) method. Here, Al_2O_3 nanoparticles at different loadings were added to study its effect on the mechanical properties. On top of that, blending of ECJO with synthetic resins was also done. All samples were later subjected to curing process for analysis. FTIR analysis has proven that CJO has been successfully converted to ECJO. Furthermore, addition of nanoparticles was found to effectively improve the mechanical properties of the specimens. 1wt% addition of Al_2O_3 , yielded the highest tensile stress of 29.37 ± 2.00 MPa with a modulus of 840.80 ± 124.53 MPa compared to the unmodified synthetic resin's tensile properties and elastic modulus of 41.42 ± 6.55 MPa and 1184.44 ± 502.54 MPa respectively. Further blending and addition nanoparticles causes decrease in the mechanical properties with tendency of agglomeration. The optimum sample of 25% ECJO blend, 1 wt% nanoparticles gave a glass transition temperature of 37.95°C. In short, the act of blending and addition of nano-fillers has improved the characteristic of ECJO for adhesive applications successfully although more investigation must be done to get the right formulation in order to compete with the petroleum-based counterparts.

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Author contribution

All authors contributed equally to this work.

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Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

Conflicts of interest

The authors declare that there is no conflict of interest.

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