Pertanika J. Sci. & Technol. 30 (3): 2127 - 2138 (2022)

PERTANIKA JOURNALS

SCIENCE & TECHNOLOGY

Journal homepage: http://www.pertanika.upm.edu.my/

Synthesis, Characterisation, and Electrochemical Impedance Spectroscopy Study of Green and Sustainable Polyurethane Acrylate from Jatropha Oil Using a Three Step Process

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ABSTRACT

Bio-based polymer is a promising candidate to substitute conventional petroleum-derived polymer as it is sustainably produced from renewable resources, which helps reduce the production process' carbon footprint. It also helps reduces humankind's dependability on fossil fuel-based feedstock. In this work, a sustainable jatropha oil-based polyurethane acrylate (PUA) was successfully prepared and synthesised using a 3-steps process; epoxidation (formation of an epoxy group), hydroxylation (addition of–OH group to opened ring), and acrylation (addition of acrylate group into polyol). The yellowish PUA prepared has a gel consistency, which is sticky and slightly runny. The PUA was characterised by using wet chemical tests such as oxirane oxygen content (OOC), acid value (AV), hydroxyl number (OHV) and iodine value. OOC value for the PUA synthesised was 4.23 % at the 5 hr reaction time. At the same time, the Epoxidised

ARTICLE INFO

Article history:
Received: 07 August 2021
Accepted: 02 November 2021
Published: 25 May 2022

DOI: https://doi.org/10.47836/pjst.30.3.21

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ISSN: 0128-7680
e-ISSN: 2231-8526

jatropha oil (EJO) used to prepare polyol records a hydroxyl number of hydroxyl 185.81 mg KOH/g and an acid value of 1.06. The polyol prepared was mixed with 2, 4-toluene-diisocyanate (TDI) and Hydroetyhlmethacrylate (HEMA) to produce PUA. The PUA was characterised by thermogravimetry analysis (TGA) and electrochemical impedance spectroscopy (EIS). TGA analysis shows that the polymer is stable up to 373 K, whereas the EIS analysis records an ionic conductivity of $(5.60\pm0.03) \times 10^{-6}$ S cm^{-1}. This polymer's great thermal stability properties make it suitable for outdoor application where high temperature due to sun exposure is common. Furthermore, PUA prepared gel-like properties to make it a suitable candidate for preparing a gel polymer electrolyte.

Keywords: Jatropha oil, polyurethane acrylate

INTRODUCTION

Polymer is a material that consists of a large molecule made up of many repeating subunits. Because of this, polymers can be chemically engineered with desired properties to meet the basic needs of society (Ahvazi et al., 2016).

In this study, the polymer should have excellent chemical, electrochemical, mechanical, photochemical, and thermal properties to be suitable for being used as an electrolyte host polymer. Ideally, the polymer should contain electron donor groups such as O, NH, CN, & F. This is important as the metal cation can form coordinate bonds with electron donor groups, allowing the polymer to dissolve iodide salts readily.

The polymer can be classified into two types: petroleum-based polymer and bio-based polymer. Petroleum-based polymers are currently being used extensively in our daily life. However, due to its environmental and sustainability issue paired with unstable oil prices, the bio-based polymer was introduced as a promising candidate to replace petroleum-based polymer (Mangaraj et al., 2019; Mohanty et al., 2005; Nagalakshmaiah et al., 2018; Siracusa et al., 2008). Bio-based polymers are derived mainly from plants. It makes biopolymers sustainable to produce, environmentally friendly and great availability (Mohiuddin et al., 2017; Sharmin et al., 2015). Examples of bio-based polymers are cellulose (Chua et al., 2020; Du et al., 2019), chitosan (Ogino et al., 2020; Zulkifli et al., 2020), and starch (Lobregas & Camacho, 2019; Tiwari et al., 2019).

Vegetable oils such as palm oil (Adam et al., 2020; Daud et al., 2015), soybean (Huo et al., 2019; Nan et al., 2020), castor (Ibrahim et al., 2015; Ibrahim et al., 2018), neem oil (Desappan et al., 2019) and jatropha oil (Ling et al., 2019; Sammaiah et al., 2014) are suitable to be used in preparing a green and sustainable bio-based polymer. In this work, jatropha oil was selected as starting material. Jatropha oil has unique properties, making it suitable to be used as a feedstock in making a bio-based polymer. Jatropha oil contains a toxic phorbol ester group, making it a non-edible oil (Hazmi et al., 2013). As the oil is

non-edible, a future increase in demand for jatropha oil will not affect the prices of food items. *Jatropha Curcas*, the plant that produces jatropha oil, is also easily cultivated in harsh environments which are not suitable for food cultivation.

Jatropha oil was used to prepare polyol ($R'-(OH)_n$), which will then be used in producing polyurethane acrylate (PUA). PUA is considered a unique polymer consisting of soft and hard segments arranged in an alternating repeating pattern within the same macromolecular chain (Unal et al., 2005). The soft segment of the PUA polymer can act as a solvent to dissolve iodide salts more readily to produce an electrolyte. In contrast, the hard segments help to maintain the electrochemical stability of the gel polymer electrolyte in electrochemical devices. Therefore, PUA polymer is a suitable candidate for an electrolyte system as a host polymer (Su'Ait et al., 2014).

MATERIALS AND METHODS

Material

Jatropha oil (JO) was procured from a local supplier named Bionas Biofuel Sdn Bhd, Malaysia. Formic acid (99 % purity), sulfuric acid (H_2SO_4), N, N-dimethylformamide (DMF), potassium hydrogen phthalate, potassium hydrogen and phenolphthalein were obtained from R&M Chemical, Malaysia. Methanol (CH_3OH) was purchased from HmbG® Chemical, Malaysia. Aqueous hydrogen peroxide (purity 30 %) was sourced from Merck, Germany. Technical grade 2-Hydroetyhlmethacrylate (HEMA) with 80 % purity, chlorobenzene, crystal violet, toluene, phthalic anhydride, pyridine, and sodium thiosulphate was procured from Sigma Aldrich, Germany. In addition, 2,4-toluene-diisocyanate (TDI) with 80 % technical grade, hydrogen bromide and glacial acetic acid were purchased from Acros Organics, New Jersey. Sodium hydroxide, starch solution and cyclohexane were supplied from Pubchem.

Preparation of Polyurethane Acrylate (PUA)

There are three steps in the synthesis process of PUA: the epoxidation, hydroxylation and introduction of the acrylate group in the urethanation process (Chai et al., 2020; Ling et al., 2019). For the epoxidation process, a mixture of 200 g of Jatropha oil and 23.32 g of formic acid was stirred using an overhead stirrer with a temperature of 40 °C for 10 min. Then, 146.52 g of 30 % hydrogen peroxide was added dropwise into the mixture via a dropping funnel. The mixture was stirred at 60 °C for 6 h after adding hydrogen peroxide. Every 1 h interval, a small amount of mixture was taken out and analysed for oxirane oxygen content (OOC) following the methodology described by ASTM D1652-97 Method A standard. 6 h later, the mixture obtained was then transferred into a separating funnel. It was left to cool to room temperature naturally. The settled aqueous layer was discarded, and the remaining acid was rinsed away with slightly warm distilled water several times

until a clear yellowish Epoxidised Jatropha Oil (EJO) was produced. From there, 0.6 g of EJO was used to test for Iodine Value (IV) according to the method described by Ainie et al. (2004).

For the hydroxylation process, 133 g methanol, 0.93 g sulphuric acid and 15 g of distilled water were pre-mixed into a three-neck round bottom flask at 50 °C for 15 min. Next, 150 g of EJO was added slowly to the three-neck round bottom flask. The mixture was then heated gradually to a temperature of 65 °C and stirred continuously for an additional 30 min. Finally, the mixture was transferred to a separating funnel and was left to cool naturally back to room temperature. The aqueous layer was removed, and the residue was then rinsed with slightly warm distilled water several times until it was clear. The excess distilled water and methanol were removed using a rotary evaporator, and polyol with a clear golden colour was successfully synthesised. The polyol sample was tested for Hydroxyl Value (OHV) and Acid Value (AV) using ASTM D 4274-99 in Method C and ASTM D4662-03 Method A, respectively.

In order to prepare PUA, the polyol was synthesised earlier, and DMF solvent was stirred using an overhead stirrer at 60 °C at 500 rpm. Then, TDI was added dropwise into the mixture and was stirred for 2 h at 80°C. After that, the mixture's temperature was then cooled to 40 °C, and HEMA was added slowly into the mixture. After adding HEMA, the mixture was heated up to 70 °C for another 1 h. A 25 mL of DMF was gradually added to produce PUA during the process. It was being done to lower the overall viscosity of the PUA polymer. The PUA synthesised were then stored in the desiccator properly to be used in the future (Figure 1).

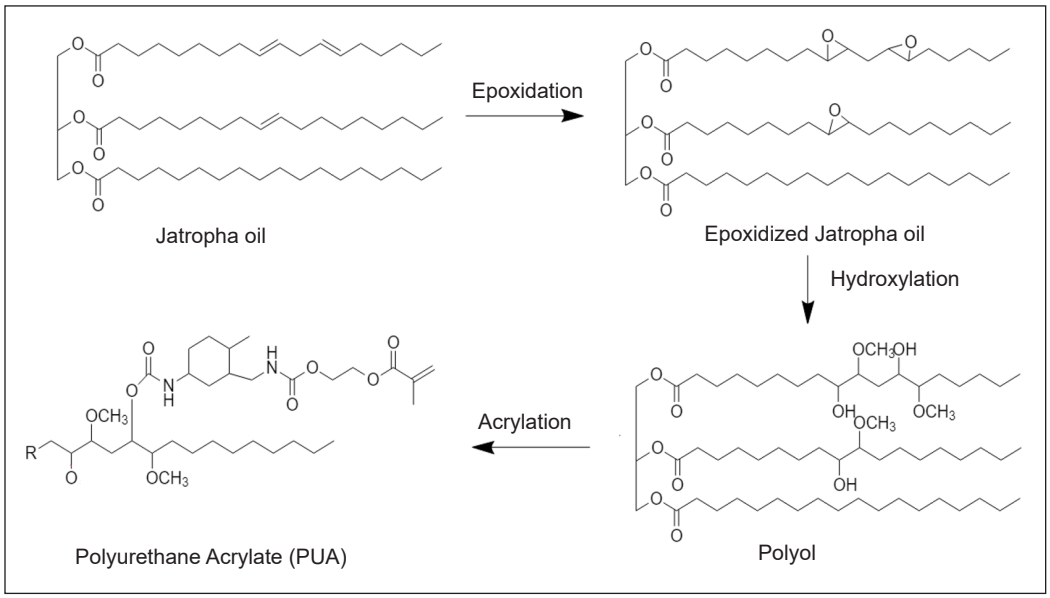

Figure 1. Reaction diagram

Thermal Gravimetric Analysis (TGA)

The PUA gel polymer electrolyte's thermal stability was analysed using TG Analyzer, Perkin Elmer TGA7. TGA was used to measure the PUA weight change as a function of temperature. The PUA gel polymer electrolyte was heated from 298 K to 873 K with a constant heating rate of 283 K/min under a nitrogen atmosphere.

Electrochemical Impedance Spectroscopy (EIS)

HIOKI IM3570 Impedance Analyser obtained the PUA gel polymer electrolyte impedance. The measurement was conducted in the frequency range between 50 Hz to 1 MHz at room temperature (298 K). The PUA was filled in a coin cell with both electrodes being stainless steel. From the Nyquist plot obtained, the value of ionic conductivity can be calculated using Equation 1:

$$\sigma = \frac{t}{A \times R} \quad (1)$$

Here, t is the sample thickness (0.26 cm), A is the electrode/electrolyte contact area (2.01 cm^2), and R is electrolyte bulk resistance.

RESULTS AND DISCUSSION

Chemical Properties

Oxirane Oxygen Content Values (OOC) and Iodine Value. OOC is used to determine the double bond conversion value in the vegetable oils into epoxy rings or three-membered rings () (Hernández-Cruz et al., 2021; Saurabh et al., 2011). During the epoxidation process, formic acid and hydrogen peroxide were used because formic acid plays an oxygen carrier role, while hydrogen peroxide is an oxygen donor. Therefore, formic acid is responsible for facilitating the transfer of oxygen from hydrogen peroxide to react with a double bond in the unsaturated fatty acid of the Jatropha oil in the formation of epoxy rings.

The EJO sample was tested hourly to monitor the OOC level during the epoxidation process. Figure 2 and Table 1 show that the OOC level increased from (3.85±0.030) % mol^{-1} at 1 h until it reached the maximum level, (4.23±0.015) % mol^{-1} at 5 h. Beyond 5 h, the OOC level decreased to (3.92±0.035) % mol^{-1}. Furthermore, Hazmi et al. (2013) reported that the produced EJO had an OOC value of 3.67 % to 3.89 %. Another example of epoxidation reaction was mentioned by Meyer et al. (2008). The OOC value achieved was 4.75 %, and the time taken was about 10 h. Although it had a higher OOC value, the time taken was prolonged. The OOC value recorded in this work is on par with previous research as it was able to achieve an OOC value of 4.23 % with a shorter time of up to 5 h (Hazmi et al., 2013; Meyer et al., 2008).

Based on the OOC value observed, the relative conversion to oxirane can be calculated as below (Goud et al., 2010):

$$Relative\ Conversion\ to\ Oxirane = \frac{OOC_{exp}}{OOC_{theo}} \times 100\% \quad (2)$$

OOC_{exp} is the experimental OOC value, and OOC_{theo} is the theoretical OOC value (6.15 %).

Based on Equation 2, the relative conversion to oxirane calculated increases from 62.60 % to 68.70 % at the 5th hour. It is desirable as higher relative conversion to oxirane will allow the OH group to form more sites. In addition, it is due to the oxirane ring being a favourable site for nucleophilic attack by the hydroxyl group via ring-opening reaction. The hydroxyl group can then be attached to an aliphatic fatty acid chain to form polyol during hydroxylation (Mohamed et al., 2020).

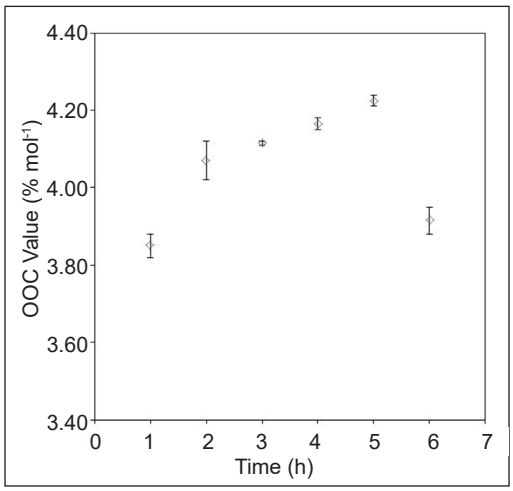

Figure 2. OOC value (%) for 6 h

Table 1
Parameters of OOC value and relative conversion to oxirane for 6 h

Time (h)	OOC value (%)	Relative Conversion to Oxirane (%)
1	3.85	62.60
2	4.07	66.18
3	4.12	66.91
4	4.17	67.72
5	4.23	68.70
6	3.92	63.66

Furthermore, the iodine value method evaluated the sample's degree of unsaturation (C=C). Jatropha oil had an iodine value of 101.25 mg I_2 /g. It was because jatropha oil has a high content of unsaturated fatty acids such as palmitic acid, C16:0 (16 %), stearic acid, C18:0 (6 %–7 %), oleic acid, C18:1 (42 %–43.5 %), linoleic acid, C18:2 (33 %–34.5 %) and linoleic acid, C18:3 (0. 8%) (Akbar et al., 2009; Amri et al., 2021). After completing the epoxidation process, EJO presents 5.60 mg I_2 /g for iodine value. The epoxidation process caused this decrease in iodine value.

Hydroxyl Value (OH Value) and Acid Value (AV). In this work, the OH value was carried out to determine the free content of the hydroxyl group that is present in the polyol by using phthalic anhydride pyridine (PAP) at a temperature of 115 °C for 1 h. For the hydroxylation process, the hydrogen atom from the acid alcohol will react with the oxygen atom from the symmetric ether (C-O-C) to form a hydroxyl group, producing polyol. The OH value calculated was 185.81 mg KOH / g. Saalah et al. (2015 & 2021) listed that the

Jatropha oil-based polyol had recorded an OH number of 138 mg KOH / g, while Hazmi et al. (2013) proved that the hydroxyl number achieved by jatropha oil-based polyol was in the range of 171 to 179 mg KOH / g with functionality 5.1 until 5.3. Another example of epoxidation reaction was mentioned by Mudri et al. (2020). They reported that OHV for their Jatropha oil-based polyol (JOL) was 149.44 mg KOH / g. Based on the results cited above, the OH value obtained in this work is higher.

A higher OH value indicated the increase of bond strength as a high crosslinking structure was formed (Park et al., 2020; Somani et al., 2003). Polyol with a high OH value will have good mechanical properties (Wang et al., 2009; Zhang et al., 2017).

Other than that, the acid value was tested on the polyol sample. First, the acid value was calculated to determine the free fatty acid, such as the acidic residue groups in the polyol. The polyol sample recorded (1.06±0.11) mg KOH/g acid value. This low acid value will be able to prevent the corrosion of electrodes in dye-sensitised solar cell applications.

Thermal Gravimetric Analysis (TGA)

TGA was used to measure the changes in weight as the sample was gradually heated. The polymer electrolyte must be heat resistant and must be able to resist a temperature of more than 373 K to achieve the stability of the electrochemical devices. This polymer electrolyte will be applied in the DSSC application, which was agreed upon by Holdt and Kraan (2011). Figure 3 shows the TGA thermogram and DTG curves of pure PUA in the temperature range of 300 K to 873 K. As the temperature increases, it will liberate some moisture and DMF solvent (Fu et al., 2020). It can be observed in the temperature range of 303 K and 500 K. It was deduced that the boiling point of DMF solvent was 425 K–427 K (Adachi & Sakka, 1988; Wannatong et al., 2004). Therefore, DMF was added during the synthesis process of PUA. Furthermore, it showed degradation of the hard segment block of urethane linkage at the temperature of 500 K and 620 K (Adam et al., 2020; Mendes-Felipe et al., 2020; Saalah et al., 2015; Wei et al., 2018). Based on Figure 3, it can be seen that a maximum peak of thermal degradation happened at 620 K to 780 K with 33 % of weight loss. It was caused by the degradation of a soft polyester segment (Adam et al., 2020; Su'Ait et al., 2014; Wei et al., 2018). Thus, Figure 3 proves that this PUA sample is suitable for DSSC application as it can endure at temperatures exceeding 423 K (Fu et al., 2020).

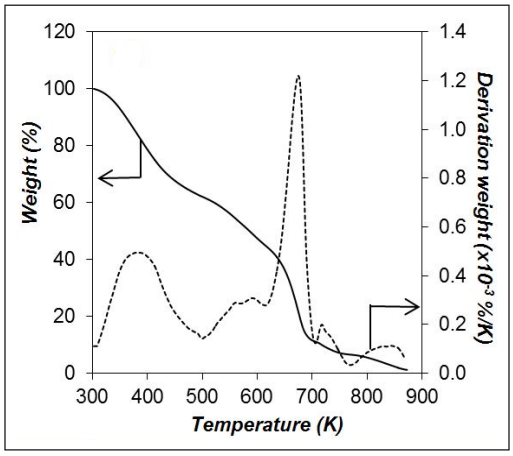

Figure 3. TGA analysis of PUA

Electrochemical Impedance Spectroscopy (EIS)

In this work, PUA is used as a medium for charge transport. In order to determine its properties, ionic conductivity becomes a main concern in the polymer electrolyte. Figure 4 shows the Nyquist plot of pure polyurethane acrylate at room temperature. Based on the Nyquist plot in Figure 4, it presented only a semicircle due to the bulk property of the material. Besides that, the R-value of the pure PUA determined from the Nyquist plot was 29400 Ω. With the R-value obtained, the ionic conductivity of the pure PUA can be calculated. The pure PUA recorded an ionic conductivity of $(5.60\pm0.03) \times 10^{-6}$ S cm^{-1}. This observation shows the ionic conductivity of the polymer to be better than previous work done by Rayung et al. (2019 & 2020). Rayung et al. (2019 & 2020) reported that their samples only managed to obtain an ionic conductivity value of 1.09×10^{-8} S cm^{-1}. The ionic conductivity in this work is two orders of magnitude higher than in previous studies (Rayung et al., 2019; Rayung et al., 2020).

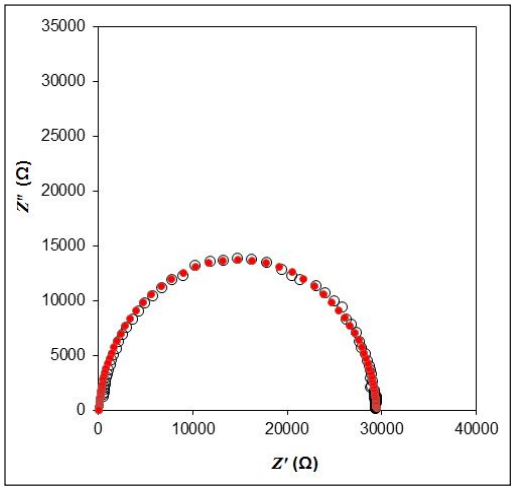

Figure 4. Nyquist plots of pure PUA at ambient temperature (o corresponds to experimental points and • corresponds to fitted points)

CONCLUSION

Jatropha oil is a suitable candidate for preparing a sustainable biopolymer polyurethane acrylate electrolyte. TGA analysis shows the high thermal stability of the polymer, which makes it a great candidate to be used as a host polymer in outdoor dye sensitised solar cells applications. Furthermore, EIS analysis of the polymer prepared shows a relatively good ionic conductivity value which indicates that it is a good starting material to be used in electrochemical applications.

ACKNOWLEDGMENTS

The authors acknowledge with gratitude the Universiti Putra Malaysia for IPS Grant (GP-IPS/2019/9682100).

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