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Thermal Behavior of Modi?ed Thermoplastic Starch (TPS) Synthesized from Sago (Metroxylon Sagu) with Diphenylmethane Diisocyanate and Castor Oil Rozanna Dewi, Nasrun Ibrahim and Novi Sylvia Chemical Engineering Department, Universitas Malikussaleh, Reuluet, Kecamatan Muara Batu, Kabupaten Aceh Utara, Provinsi Aceh, Indonesia Dahlan Abdullah Informatics Engineering Department, Universitas Malikussaleh, Reuluet, Kecamatan Muara Batu, Kabupaten Aceh Utara, Provinsi Aceh, Indonesia Medyan Riza Chemical Engineering Department, Universitas Syiah Kuala, Darussalam, Kecamatan Syiah Kuala, Kota Banda Aceh, Provinsi Aceh, Indonesia Abstract Purpose – The purpose of this research is to blend modi?ed thermoplastic sago starch (TPS) through in-situ instrument by responding sago starch with diphenylmethanediisocyanate (MDI) and castor oil at the same time, bringing about a more homogenous and ?ner-sized polyurethane prepolymer (PUP).

Design/Methodology/Approach – The methods used were Thermal Gravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC) for thermal characterization and stability of PUP, modi?ed TPS non- extracted and extracted with toluene and water. Findings – TGA test results presented shows that PUP begins to decompose thermally at a temperature of 300–500 °C.

Weight loss occurs rapidly between these temperatures and is completely discharged at a temperature of 500°C, which is called weight loss transition. Research Limitations/Implications – When removed with toluene and a water dissolvable, the softening point and dormant warmth of combination marginally diminished; be that as it may, it is as yet higher than the first estimation of sago.

In terms of thermal stability, modi?ed TPS decomposes and loses weight at 150–200 °C in small quantities, continues with weight loss rapidly, and is completely discharged at 500°C. The thermal stability is considered high; thus, modi?ed TPS application can be varied. Practical Implications – DSC analysis and TGA shows that modi?ed TPS has good thermal characteristics and thermal stability. Modi?ed TPS has a melting point of 104.69°C, and the latent heat of _Thermal Behavior of Modi?ed Thermoplastic Starch 387 __

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Proceedings of MICoMS 2017 388 _fusion (DH) is 234.27 J/g.

This value is close to the PUP melting point and latent heat of fusion, which reveals the formation of cross-link between the starch and PUP. Keywords Sago, modi?ed thermoplastic starch (TPS), castor oil, Methylene Diphenylene Diisocyanate, (MDI), thermal characteristics All papers within this proceedings volume have been peer reviewed by the scienti?c committee of the Malikussaleh International Conference on Multidisciplinary Studies (MICoMS 2017).

Introduction The basic materials used today for plastic are polypropylene and polyethylene. The waste produced is high and harmful for the environment since it cannot be degraded naturally. Eventhough the plastic waste can be reused, and open conversation related with the degree of security and wellbeing for the client, particularly since the issuance of Peraturan Kepala Badan Pengawasan Obat dan Makanan RI No. HK 00.05.55.6497 Tentang Bahan Kemasan Pangan (Head of Drug and Food Control Body Regulation on Food Packaging Materials) on 2008, which disallows utilizing of reused plastics for food bundling (Pudjiastuti et al., 2012).

Asia is the world's biggest buyer of plastic; it assimilates about 30% of worldwide plastic utilization, and around 100 million tons of plastic is created on the planet for different modern divisions (Dayanti, 2006). Alongside the expanded mindfulness for ecological protection, the requirement for biodegradable plastic materials has expanded.

In 2010, the production of biodegradable plastics was projected to reach 1.2 million tons (Dayanti, 2006). Starch has been generally utilized as crude material for biodegradable plastic because of its biodegradability, sustainably, and accessibility. Plastic arranged from starch, with its low measures of water, is fragile.

To lessen the fragility, starch is plasticized with hydrophilic plasticizers, for example, glycerol, and is softened for getting ready thermoplastic starch (TPS). Following a while, glycerol-plasticized TPS becomes fragile on the grounds that glycerol movement from starch network cooperates with starch by non-covalent hydrogen holding prompting stage partition. Joining a ?exible effect modi?er to the framework starch with the covalent bonds can forestall it (Wu et al., 2008).

With the current available modi?ers, isocyanate bunches have high movement to respond with the hydroxyl gathering of starch. Polyurethane prepolymer (PUP) is utilized to harden starch. Polyol delicate sections in polyurethane (PU) is connected to starch framework by urethane linkage and assumes a job as an effect modi?er.

Security of the earth should be possible by utilizing polyol from inexhaustible materials, for example, vegetable oil (Lu et al., 2005). Among numerous sorts of plant oils, castor oil has three hydroxyl gatherings and is a contender to integrate poliueratan (Carme Coll et al., 2008). Ferrer et al. (2007) has characterized the polyurethane network synthesized from vegetable-based polyol and compared it to polyurethane network from petroleum-based.

Polyurethane network can be synthesized fromvegetable-based polyol and petroleum-basedpolyol. The purpose is similar, to react with isocyanate and form polyurethane. The different is only on the biodegradability of polyurethane formed, where the vegetable based polyol will make the Polyurethane biodegradable in nature (Ferrer et al., 2007). Wu et al.

(2008) has conducted a research on synthesizing modi?ed TPS using corn starch with PUP from diphenyl methanediisocyanate (MDI) and polyols produced from castor oil. This modi?cation produces micro particle ?llers, e.g., polyurethane prepolymer (PUP) and will consequently resulting micro composite sago starch material. The process Wu conducted strengthened thermoplastic corn starch using PUP that binds to the starch matrix through the urethane bond.

In this case, the PUP mixed into the starch matrix as ?ller after synthesized separately and already forming micro particle PUP (Wu et al., 2008).

In this study, we want to synthesize modi?ed TPS through an in-situ mechanism by reacting sago starch with MDI and castor oil simultaneously, resulting in a more homogenous and ?ner-sized polyurethane prepolymer (PUP).

Modi?ed TPS structure, forming mechanism, properties, and biodegradability have been published previously (Dewi et al., 2014). The thermal behaviours of modi?ed TPS were characterised through DSC analysis and TGA, and the result will be discussed in this paper. Experimental Materials Sago Starch, Castor oil, DiphenylmethaneDiisocyanate, sorbitol, and glycerol plasticizer.

Synthesis of modi?ed TPS Sago (31 g) was placed into an Erlenmeyer ?ask and blended in with 155 ml of water. Sago is warmed and mixed until cooked to become gel at gelatinization temperature 70 °C for around 30 minutes. After the gel is nearly framed, castor oil (4 g) and MDI (3 g) are poured straightforwardly and mixed along with enthusiastically blending for a couple of moments.

Sorbitol 7% (14 g) is likewise included. The blend is poured onto a sheet of glass for dainty printing. Film was dried in encompassing conditions for 24 h. After this, it was expelled from its projecting and slice to ?t the testing prerequisites. As a correlation, polyurethane prepolymer (PUP) was additionally created by responding castor oil (15 g) and MDI (13 g).Once the mixture is homogenous, it is printed and dried to form the PUP ?lm.

Analysis conducted The tests used were Thermal Gravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC) for thermal characterization and stability of PUP, modi?ed. TPS non- extracted and extracted with toluene and water are the type or variant of samples tested with DSC and TGA.. Result and discussion The melting point is a condition where the sample temperatures increases, so the sample state change from solid into liquid.

In contrast to metals, plastics generally do not consist of a speci?c melting point. The value of melting point is needed to determine the product's process and application conditions. The plastic would be good as packaging material or not and survive at a certain temperature in accordance with the plastic durability to be determined with thermal stability analysis. DSC analysis was conducted for the sample PUP, modi?ed TPS, modi?ed TPS extracted with toluene and water.

Images of DSC test results are shown by Figures 1–3. The DSC for the PUP test result showed a melting point of 105.55 °C and a latent heat of fusion (DH) is 224.38 J/g; for the modi?ed TPS, the melting point is 104.69 °C and the DH is 234.27 J/g. The modi?ed

TPS softening point was gotten nearly similarly as the PUP which demonstrates the improvement of cross-interface among starch and PUP, when contrasted with the sago starch which is just around 70°C.

The expansion of starch grid into the PUP expanded the dissolving purpose of the modi?ed TPS and demonstrated the event of cross-joins. The higher liquefying point shows a developing number of cross-joins happened. It is in?uenced by hydrogen bonds contained in plastic. More hydrogen bonds in ?lm mean the higher softening point will be because of vitality required to break bonds.

The melting temperature and latent heat of modi?ed TPS extracted with toluene and water diminished. Modi?ed TPS extracted with toluene showed a melting point of 98.23 °C _Thermal Behavior of Modi?ed Thermoplastic Starch 389 __

Proceedings of MICoMS 2017 390 Figure 1. DSC Analysis Modi?ed TPS Figure 2.

DSC Analysis for Modi?ed TPS Extracted with Toluene _ / and a DH of 178.76 J/g; for TPS extracted with water, the melting point is 74.86 °C and the DH is 87.74 J/g. The decrease in the melting point and DH is due to a certain partial amount of castor oil and MDI dissolved in the toluene and due to sago starch dissolved in water solvent.

__ __ Figure 4. Thermal Decomposition of PUP

Proceedings of MICoMS 2017 392 Figure 5.

Thermal Decomposition of Modi?ed TPS _ / In term of thermal stability, TGA test results showed that PUP decomposes thermally at a temperature of 300–500 °C as show n in Figure 4. Weight loss occurs rapidly between these temperatures and is completely discharged at a temperature of 500 °C, which is called weig ht loss transition (Figure 5).

The modi?ed TPS decomposes and loses weight at 150–200 °C in small quantities, continues with weight loss steeply, and is completely discharged at 500 ° C. Modi?ed TPS loses weight faster than PUP because of the weight loss occurring in the ?rst transition phase, which means the starch component has lower thermal stability compared with PUP.

Considering the high temperature range where the modi?ed TPS decomposed until it is fully decomposed, the application of modi?ed TPS can be varied within that temperature range. Having said this, there is a need to combine thermal behavior with the mechanical characteristic that has been published previously to ?t the best application (Rozanna et al., 2014). Conclusion From DSC analysis and TGA conducted, modi?ed TPS has good thermal characteristics and thermal stability.

Modi?ed TPS has a melting point of 104.69 °C, and the DH is 234.27 J/g. This value is close to the PUP melting point and the DH, which reveal the formation of cross-link between starch and PUP. When extracted with toluene and water solvent, the melting point and DH slightly decreased; however, it is still higher than the original value of sago.

In term of thermal stability, modi?ed TPS decomposes and loses weight at 150–200 °C in small quantities, continues with weight loss rapidly, and is completely discharged at 500 °C. The thermal stability is considered high; thus, the modi?ed TPS application can be varied.

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