THE MECHANICAL PROPERTIES OF GREEN POLYBLEND BASED ON WASTE POLYPROPYLENE FILLED IN VARIATION PARTICLE SIZE NATURAL FIBER AND INITIATOR CONCENTRATION

Neng Sri Suharty¹, Basuki Wirjosentono², Maulidan Firdaus³

^{1,3}Faculty of Mathematics and Natural Sciences, Sebelas Maret University, Kentingan, Surakarta, Indonesia

²Faculty of Mathematics and Natural Sciences, University of North Sumatera, Padang Bulan, Medan, Indonesia

Email: nsuharty@yahoo.com

ABSTRACT

Synthesis of green polyblend from waste polypropylene (recycle polypropylene, rPP) filled reinforcing husk rice powder (HR) in weight ratio rPP/HR (8/2) using multifunctional acrylic acid (AA) processed reactively in internal mixer had been carried out. To understand the effect of initiator concentration, some concentration variations of benzoyl peroxide (BPO) initiator had been done in 5 levels (0; 0.02; 0.05; 0.07; and 0.10 phr). Based on the mechanical properties (tensile strength, TS) and the melt flow index (MFI) in thermoplastic condition, it was found that the best concentration was 0.02%. To study the effect of particle size, four level variations (40; 70; 100 and 120 mesh) of the HR particle size using 0.02 phr BPO was prepared. It has been found that decreasing the particle size (40 to 120 mesh) decrease the MFI accordance with ASTM and increase the TS value. To understand the correlation between particle size and biodegradability of the green polyblend, firstly, it was performed water absorption characterization to the polyblend specimen. Water absorption (WA) studies showed an increase in water uptake with increase in particle size. Biodegradation of the green polyblend (reflected by the lost of weight, LW) was done by burying the specimen in cellulolytic bacteria enriched garbage soil for four months. The result showed that increasing the HR particle size (120 to 40 mesh) increase the biodegradation properties.

KEYWORDS: Green polyblend, polypropylene recycle, husk rice powder, biodegradation.

1.0 INTRODUCTION

Polypropylene (PP) generally utilizes as packaging materials and becomes waste when it is not used anymore. This PP waste can be used to other useful

product and will not contaminate the environment. This PP waste (referred as PP recycle, rPP) has reactive functional group >CH- which is capable to react with other reactive functional group [Cowd, 1991, Fessenden and Fessenden, 1992]. As an agrarian country, Indonesia plants paddy widely for the main food. Therefore, the husk rice (RH) waste was in a lot of quantity which can also contaminate the environment [Haygreen and Bowyer, 1996]. RH mainly consist cellulose, besides hemicellulose and lignin. Cellulose has three polar free reactive hydroxyl groups and two glycoside carbon groups. In this research, RH powder has been utilized as a reinforcing material for the synthesis of biodegradable green polyblend. Both rPP and cellulose RH have functional group which are different in its polarity [Fengel and Wagener, 1995]. To bind between cellulose and polypropylene, it needs to modify using acrylic acid (AA) as multifunctional, owning non polar vinyl group and polar reactive hydroxyl groups [Suharty, 2001]. The use of AA as a modifier will realize the formation of green polyblend. The methine group of rPP will bind with the vinyl group of AA and the hydroxyl group of AA will bind with hydroxyl group from cellulose to form ester [Suharty and Wirjosentono, 2005]. To improve the binding properties, divinyl benzene (DVB) was used as crosslinker agent. The utilization of RH as a filler can improve not only the mechanical properties of green polyblend but also the character of its biodegradability. In order to prove the nature of its biodegradability, the green polyblend were technically buried in cellulolytic bacteria enriched garbage soil [aSuharty and Firdaus, 2007, bSuharty et.al., 2007]. Monitoring were done periodically every one months with measuring the change of its weight caused by natural degradable process.

2.0 MATERIALS AND METHODS

All chemicals as starting compounds are in pro-analysis grade except mentioned, and used as such without any further purification. The rPP was obtained from waste drinking water cup. The rPP samples were prepared by washing the waste drinking cups with ethanol, and chopped in 2x2 mm. The RH used was first cleaned with ethanol, dried in oven at 40°C and ground into 40, 70, 100 mesh and 120 mesh particle size. Characterization of starting materials as well as new material produced were performed using: 1) Atlas Melt Flow Indexer (MFI) in order to determine the melt flow index of bio-composite in accordance with ASTM 1238; 2) Paul Weber Machinen-u-Aparatebau D-7064 Rhemshalden-Brunbach hot press was utilized to prepare specimens; 3) Tensometry (MPG SC-2 DE) was used to determine tensile strength, TS, in accordance with ASTM D638; 4) Shimadzu FTIR-8201 PC was used to determine functional group; 5). The water absorption of polyblen were determined following ASTM D 570-95; 6) Burial method in cellulolytic bacteria enriched garbage soil for four months to perform biodegradability test; and 6) morphology analysis were done with SEM JSM-35C (2500 times magnification).

2.1 Synthesis of PP green polyblend using filler RH fibre

The synthesis of PP green polyblend were conducted by laboplastomill internal mixer, reactively process at 180°C for 10 minutes. The mixture composition of process are as follow: weight ratio rPP/RH=8/2, BPO initiator concentration: 0.02; 0.05; 0.07 and 0.10 phr, 10% AA to RH weight, DVB crosslinker agent (5% from AA weight), and some variation of RH particle size: 40, 70, 100 and 120 mesh. The resulting green polyblend rPP/DVB/AA/RH were then analysis the MFI, TS, functional group, WA, LW and morphology.

3.0 RESULT AND DISCUSSION

3.1. Characterization Functional Group of Starting Materials

Each starting material compound used for the synthesis of bio-composite was spectroscopically characterized by FTIR. The FTIR spectrum of fresh AA in neat liquid shows the absorption at 1728 cm⁻¹ (C=O unsaturated ester group), 1636 and 1411 cm⁻¹ (>C=CH2 for acrylic double bond). The FTIR spectrum of rPP film shows absorption at 2890 and 972 cm⁻¹ indicate >CH- methine group of PP; absorption at 2723 and 1458 cm⁻¹ refer to -CH2- of PP. The FTIR spectrum of RH in KBr pellet shows broad absorption at 3448 from -OH cellulose hydrogen bonding; 2922 cm⁻¹ (-CH₂) ; 1651 cm⁻¹ (>C=C< aromatic); 1423 cm⁻¹ (-CH₂. stretching); 1049 cm⁻¹ (-C-O-C- stretching of cellulose) [^aSuharty and Firdaus, 2007].

3.2. Characterizations of Green Polyblend Functional Group

Furthermore, the formed green polyblend was then analyzed on its functional group in Figure 1. The spectrum green poliblend above shows the loss of methine group of PP, the decrease of hydroxyl group cellulose of RH, the friction of acrylic acid carbonyl group at 1728 cm⁻¹ to be an ester carbonyl group at 1732 cm⁻¹. The move of position carbonyl group is caused by esterification between wood cellulose and carboxylic group of AA. The disappear of vinyl group of AA at 1636 cm⁻¹, due to the double bond of vinyl binds with methine group of PP, [^aSuharty and Firdaus, 2007, ^bSuharty *et.al.*, 2007, ^cSuharty *et.al.*, 2007].



FTIR Spectrum of green polyblend rPP/DVB/AA/RH, RH particle size 120 mesh, with ratio weight rPP/RH = 8/2, BPO concentration 0.02 phr, AA 10% from RH, DVB 5% from AA, in film

3.3. Characterization of Green Polyblend's Mechanical Properties

Parameters used to determine the optimum condition process were referred to the best tensile strength property of green polyblend and the MFI value still in thermoplastic viscosity. The compatibility of rPP and RH can be increased by using multifunctional compound AA and DVB crosslinker agent. The presence of initiator can initiate the reaction between PP and cellulose of RH, on the other hand the bigger concentration can degrade the matrix of polymer by increasing the MFI values.



FIGURE 2

MFI and TS values of green polyblend (rPP/DVB/AA/RH), RH particle size 70 mesh, weight ratio of rPP/RH=8/2, AA 10% from RH, DVB 5% from AA, with some variation of BPO concentration: 0.02; 0.05; 0.07 and 0.10 phr

The polyblend were processed in optimum condition from some variation ratio of BPO concentration and in ratio rPP/RH=8/2 and RH particle size 70 mesh it was shown that the highest value of TS values of rPP/DVB/AA/RH were 0.02 phr, as shown in Figure 2. The MFI of green polyblend in this condition decreased 10% compared to that of in BPO concentration 0.05 phr. On the other hand, the tensile strength of green polyblend in optimum condition increased 44% compared to that of in BPO concentration 0.05 phr, see Figure 2.





MFI and TS value of green polyblends (PPR/DVB/AA/RH), in weight ratio of PPR/RH=8/2, with BPO concentration 0.07 phr, AA 10% from RH, DVB 5% from AA, with some variation of RH particle size: 40, 70, 100 and 120 mesh

The optimum condition of preparation green polyblend rPP/DVB/AA/RH in weight ratio rPP/BP = 8/2 using BPO concentration 0.07 phr was in HR particle size 120 mesh. It was found that the TS values of green polyblend increased significantly up to 28% compared to that of RH particle size 40 mesh (Figure 3). On the hand the MFI values of polyblend decreased 34% compared to that of RH particle size 120 mesh was higher than 40 mesh, due to the smaller particle size the bigger surface area of RH. Thus, can make the opportunity of collision reaction increase. It will cause rising the net matrix as well as increasing the TS properties, on the other hand, it will decrease the MFI.

3.4. Degradation of green polyblend microbiologically

To understand the behavior of green polyblend in water absorption, it was measured the water absorption of polyblend by soaking the film in the aquadest for 24 hours. The presence of cellulose in the polyblend can improve hydrophylicity. It was shown that the rPP process was not absorbing water. On the other hand, green polyblend in optimum condition (RH 120 mesh and BPO 0.02 phr) absorbed water. In term of water absorption (WA) capability, the green polyblend processed using RH particle size 40 mesh was the highest one as presented in Figure 4.



FIGURE 4

Water Absorption and Lossing Weight percentages after biodegradation of rPP/DVB/AA/RH with BPO concentration 0.07 phr, in variation particle size of RH: 40 - 120 mesh

Biodegradation process was performed for 4 months in the garbage dump land. To understand the effect of biodegradation was measured the loosing weight of bio-composite after exposure [^aSuharty and Firdaus, 2007, ^cSuharty *et.al.*, 2007]. The starting material rPP processed can not degradable, because no reducing of weight after biodegradation for 4 months. The result of the weight changing of green polyblend after biodegradation can be seen at Figure 4. All of the synthesized green polyblend were reduced their weight. It means that the biodegradation process had done. The highest degradability properties of the bio-composite was the bio-composite with RH particle size 40 mesh, the loss of weight up to 9.5%, see Figure 4. These datum was supporting by the water absorbtion datum which showed the similar profile [Wirjosentono *et.al.*, 1999, Kim *et.al.*, 2005].

Visual observation the morphology of microbiologically degradation is as follows: either rPP process and green polyblend specimen before degradation was white transparent with smooth surface, but after biodegradation, the specimen changed to grey transparent with harsh surface. The photo SEM of rPP/DVB/AA/RH green polyblend specimen before degradation in 2500 times magnification is in smooth surface, but after biodegradation its turn to be brittle and the surface scraggly and holes, as can be seen in Figure 5 [^aSuharty and Firdaus, 2007, ^cSuharty *et.al.*, 2007].



FIGURE 5 (a) before biodegradation, (b) after biodegradation for 4 months.

4.0 CONCLUSIONS

The optimum BPO concentration for the preparation of green polyblend (rPP/DVB/AA/RH) with ratio rPP/RH = 8/2, was 0.02% of total weight. The best TS properties for the synthesis of polyblend (rPP/DVB/AA/RH) with ratio rPP/RH = 8/2 reactively process was in RH particle size 120 mesh. The water absorption of polyblend (rPP/DVB/AA/RH) was significantly raised for the increasing of the RH particle size up to 40 mesh. The biodegradation as represented by the loosing weight of the polyblend was increased in polyblend using bigger RH particle size up to 40 mesh.

5.0 AKNOWLEDGEMENT

Indonesian Department of Research and Technology, for the financial support in the form of Basic Research Incentive.

6.0 **REFERENCES**

- M.A. Cowd,1991, "Kimia Polimer", Translate by: H. Firman, Institut Teknologi Bandung Press, Bandung.
- R.J. Fessenden, R. F. Fessenden, 1992, "Kimia Organik", Translate by: A.H. Pudjaatmaka, Volume 2, Edition. 3, Erlangga Publisher, Jakarta.
- J.G. Haygreen and J. Bowyer, 1996, 'Hasil Hutan dan Kayu", Gadjah Mada University Press, Yogyakarta.

- D. Fengel, G. Wagener, 1995, "Kayu: Kimia, Ultrastruktur, Reaksi-reaksi", Translate by: H. Sastrohamidjojo, Gadjah Mada University Press, Yogyakarta.
- N.S. Suharty, 2001, "Reactive Processing of Hindered Phenol as Antioxidant in Polypropylene", *Prosiding Regional Conference for Young Chemist* 2001, Universiti Sains Malaysia, Penang, Malaysia.
- N.S. Suharty and B. Wirjosentono, 2005, "Impregnasi Reaktif Kayu Kelapa dengan Limbah Plastik Polistirena serta Penyediaan Komposit Polistirena Menggunakan Penguat Serbuk Kayu Kelapa", *Jurnal Alchemy*, Vol. 4 No. 2.
- ^aN.S. Suharty and M. Firdaus, 2007, "Synthesis of Degradable Bio-Composites Polystyrene Recycle Modified in Reactively Process Using Natural Fibre Filler", Prosiding International 12th Asian Chemical Congress, Kuala Lumpur, Malaysia
- ^bN.S. Suharty, B.Wirjosentono and M. Firdaus, 2007, "Pembuatan Biokomposit Degradabel Dari Polipropilena Daur Ulang Dengan Serbuk Sekam Padi Atau Serbuk Bambu", Report of Competition Research Grant XVI/1, General Higher Education, Jakarta.
- ^cN.S. Suharty, B. Wirjosentono and M. Firdaus, 2007, "Pembuatan Poliblen Degradabel Dari Limbah Kemasan Polipropilena Dengan Bahan Pengisi Serbuk Sekam Padi Dan Pemlastis Crude Palm Oil (CPO) Secara Reaktif", Report of Basic Research Incentive Under Indonesian Department of Research and Technology Year I/2, Jakarta
- B. Wirjosentono, S. Pandia, Nursyamsu Bahar, R. Dalimunthe, E. Munir, P. Guritno, and D. Y. Nasution, 1999, "Pembuatan poliblen mampu terdegradasi menggunakan teknik pengolahan reaktif poliolefin dan serbuk limbah kelapa sawit" Report of Competition Research Grant V, General Higher Education, Jakarta.
- H.S. Kim, H.S. Yang and H.J. Kim, 2005, "Biodegradability and Mechanical Properties of Agro-Flour-Filled Polibutylene Succinate Biocomposites", *Journal of Applied Polymer Science*, Vol. 97, 1513-1521.