EFFECT OF LIQUID NATURAL RUBBER ADDITION TO THE MECHANICAL PROPERTIES OF ELASTOMERIC THERMOPLASTIC / POLYETHYLENE POLYBLEND

PENGARUH PENAMBAHAN KARET ALAM CAIR TERHADAP SIFAT MEKANIK POLYBLEND TERMOPLASIK ELASTOMER / POLIETILEN

Deswita[1], Sudirman[1,2], Aloma Karo Karo[1] and Dian Iramani[3]

[1]Center for Technology of Nuclear Industry Materials-National Nuclear Energy Agency Indonesia
Gedung 71-BATAN, Kawasan Puspiptek,Serpong 15314,Indonesia
[2]Department of Chemistry, University of of Indonesian
Kampus Baru UI, Depok
[3]Centre for the Application Isotopes and Radiation Technology-National Nuclear Energy Agency Indonesia
Jl. Lebak Bulus Raya No.49, Jakarta 12070, Indonesia

E-mail: deswita@batan.go.id

Received 17 September 2012; revised 28 September 2012; accepted 23 Oktober 2012

ABSTRACT

Addition of liquid natural rubber (LNR) to elastomeric thermoplastic (ETP)/polyethylene (PE) polyblend has been done. The aim of this research is to study the effect of LNR addition to the ETP /PE polyblend using blending process. The blending process was done by mixing of ETP and PE with composition of 30% and 70% by weight respectively. LNR was added to the ETP /PE with composition of 3%, 5%, and 7% by weight, to form composite materials, refer as PLB-3, PLB-5 and PLB-7. The density of the specimen was measure by picno-meter, the mechanical properties were tested by Stograph R-1, the thermal property was analyzed by Differential Thermal Analysis (DTA) and the microstructure was observed by Scanning Electron Microscope (SEM). The result shows that the melting point of ETP/PE poly-blend increased from near 140^{o}C to near 160^{o}C with addition of LNR, but decomposition temperature decreased. Likewise, the mechanical properties of ETP/PE poly-blend exhibit the improvement after being added LNR. The mechanical properties show a rigid structure with the highest tensile strength of 191 Kg.m^{-2}, and referred as the most optimum composition ETP/PE poly-blend. This condition is reached with addition of 5% by weight LNR showing regular and homogenous microstructure. It can be concluded that the addition of LNR could improve miscibility of ETP/PE, so that a better quality was obtained.

Key words : Elastomeric thermoplastic, Polyethylene, Liquid natural rubber, Polyblend

ABSTRAK

Penambahan karet alam cair (*Liquid Natural Rubber*/*LNR*) ke poli-campuran termoplastik elastomer (*elastomeric thermoplastic ETP*) / polietilen (PE) telah dilakukan. Tujuan dari penelitian ini adalah untuk mempelajari pengaruh penambahan *LNR* ke *ETP*/PE poli-campuran menggunakan proses blending. Proses blending dilakukan dengan mencampur *ETP* dan PE dengan komposisi masing-masing 30% dan 70% berat. Kemudian *LNR* ditambahkan ke *ETP*/PE dengan komposisi 3%, 5%, dan 7%berat untuk membentuk material komposit, merujuk sebagai, PLB-3 PLB-5 dan PLB-7. Densitas spesimen diukur dengan piknometer, sifat mekanik diuji oleh Stograph R-1, Sifat termal dianalisis dengan *Differential Thermal Analysis* (*DTA)* dan strukturmikro diamati dengan *Scanning Electron Microscope* (*SEM*). Hasilnya menunjukkan bahwa titik leleh *polyblend ETP*/PE meningkat dari 140 ^{o}C ke 160 ^{o}C dengan penambahan *LNR*, tetapi suhu dekomposisi turun. Demikian juga, sifat mekanik *ETP*/PE *polyblend* menunjukkan perbaikan setelah ditambahkan *LNR*. Sifat mekanis menunjukkan struktur kaku dengan kekuatan tarik tertinggi 191 kg.m^{-2}, dan disebut sebagai komposisi *ETP*/PE *polyblend* yang paling optimal. Kondisi ini dicapai dengan penambahan 5 %berat *LNR* dengan memperlihatkan strukturmikro teratur dan homogen. Dari penelitian ini dapat disimpulkan bahwa penambahan *LNR* dapat meningkatkan *miscibility* dari *ETP*/PE sehingga diperoleh kualitas yang baik.

Kata kunci : Termoplastik elastomer, Polietilen, Karet alam cair, *Polyblend*

INTRODUCTION

Elastomeric thermoplastic (ETP) represent one of plastic polymer type made from natural rubber and have excess compared to other polymer type, namely do not need crosslink agent, more simple process, nature of better physical and recycle-able (Dahlan, Khairul Zaman A and Ibrahim 2002; Ismail and Suryadiansyah 2002; Sudirman, et al. 2000). Indonesia is one of natural rubber producer, with production of about 1.4 million ton per year. The usage of ETP have especial upon which at car, weapon and plane industry, on that account production of ETP very according to be developed in Indonesia so that can improve natural rubber added (Olk, Pena and Ralf 2004; Deswita et al., 2002).

Currently, the common uses of materials based on ETP/PE reinforced rubber, glass or plastics have broad, propen applications include boats, automobiles, baths, hot tubs, water tanks, roofing, pipes, cladding, casts, external door skins, and sport equipments sectors (Fink 2010). Even more specifically are very attractive for aircraft and aerospace structural parts. However, much of the technology is new and not presented formally in secondary or under-graduate education, and the technology of advanced composites manufacture is continually evolving (Osha, 2010).

Usually ETP are less stable to high temperature and more flexible than other plastic materials and have very low mechanical properties, which are very soft and very brittle in the dry state, so as to be used in industry, such as for cars and aircraft industry accessories are not allowed. However, the ETP is very lightweight and easy to make in a variety of forms. For this purpose, the mechanical properties of the material must be high (Envis 2003; Halimatudahliana, Nasir and Ismail 2001).

One way to improve the mechanical properties and hardness of these materials is to form composite materials. By adding other polymer/copolymer such as polyethylene (PE) which may form a ETP/PE poly-blend they can be improved. In this research, ETP/PE polyblend was added with Liquid Natural Rubber (LNR) as a compatibilizer to form ETP/PE Polyblend-LNR composite materials. In point of view, compatibility among the phases is playing an important role which can be deceived by missing a material of compatibilizer to improve adhesion among the phases. The materials of compatibilizer can be expected to improved homogenity and nature of physics as well as mechanical properties, yielded a preeminent polyblend (Deswita et al. 2008; Mahsuri, Marini and Sudirman 2005; Mashuri et al. 2008; Deswita et al. 2008; Utama et al. 1995).

This research studied the influence of addition liquid natural rubber (LNR) as compatibilizer with variation composition to poly-blend of natural rubber based ETP/PE. From this research a better ETP having nature of physical and mechanical properties is expected for materials industry.

MATERIAL AND METHOD

Material

Material for this research are polyethylene (PE) and liquid natural rubber (LNR).

Method

ETP resulted from γ-irradiation process was mixed with polyethylene (PE) with comparison % of weight 70:30. The mixing than were added by LNR with composition of 0%, 3%, 5% and 7% by weight, later they called as PLB-0, PLB-3, PLB-5 and PLB-7 respectively (Deswita et al. 2008; Halimatudahliana, Nasir and Ismail 2001). Materials containing of TPE, PE and LNR were then inserted into a Haake (RHEOMIX 3000/3010) owning capacities 250 g processing at 130^{o}C for 10 minute, for each LNR variation composition (Mahsuri, Marini and Sudirman 2005). The process was confined by pressing use a hydraulic hot press at pressure of 150 $Kg.cm^{-2}$ followed by hydraulic cold press at pressure of 16 ton.

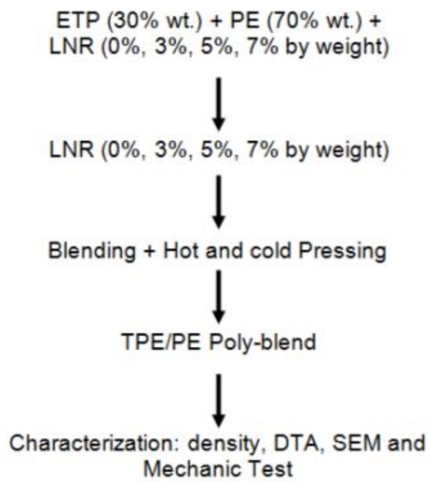

Figure 1. Synthesis and characterization of ETP/PE/LNR composite materials with LNR composition varation.

Examination of the specimen was conducted covers mechanical test using Stograph R-1 for tensile, yield strength and elongation at break. The microstructure of the specimen was observed by SEM (Scanning Electron Microscope), while the thermal property was measured by Differential Thermal Analysis (DTA). Synthesis and characterization of ETP/PE polyblend is mentioned schematically in Figure 1.

RESULT AND DISCUSSION

Thermal Properties

The thermal property of the materials was measured using Differential Thermal Analysis (DTA). The purpose of this measurement is to determine the transition temperature for the conversion of the precursor to ETP/PE polyblend phase which is demonstrated in Figure 2. Based on this Figure 2, it can be seen that there are two different endothermic peaks of each specimens.

The first endothermic peak is near 140 ^oC for origin specimen (PLB-0), near 161 ^oC for PLB-3, 163 ^oC for PLB-5, and 165 ^oC for PLB-7. These peaks give information about melting point of these materials. The second endothermic peak is near 526 ^oC for PLB-0, near 495 ^oC for PLB-3 and near 493 ^oC for PLB-5 and PLB-7, respectively. These endothermic peaks represent the decompositiontemperature of each of the specimens. It can be seen that the melting point of poly-blend materials increased with addition of LNR to 7% by weight. However, the decomposition temperature of polyblend with LNR content is lower than that of poly-blend origin.

This phenomenon indicates that the content of LNR in the ETP/PE polyblend influenced to changes in the thermal properties of both the melting point and decomposition temperature, see Figure 2. To know the mechanical properties, including yield, tensile and elongation break, the specimen were tested using Stograph R-1 apparatus.

Physical Properties

The result of density measurement is demonstrated at Table 1. It can be seen that the density of polyblend increases with increasing LNR addition. The origin sample (PLB-0) without content LNR, the density is very low only about 0.40 g.cm^{-3}, and then the density increased to about 0.75 g.cm^{-3} with increasing the content of 7% by weight LNR. The increasing of sample density can be assumed as increasing of molecular packing due to the missibility improvement.

To determine the effect of density on yield and tensile strength, all the specimens were tested the mechanical and physical properties using *Stograph* R-1 equipment. While the density of the specimen and variation of the LNR concentration was observed using SEM.

Figure 2. DTA curve from specimens PLB-0, PLB-3, PLB-5 and PLB-7

Mechanical Properties

The mechanical properties, including yield and tensile strength and also elongation break of the specimens were tested using Stograph R-1 equipment that avaliable at PTBIN-BATAN. The the mechanical test result was shown in Table 1 for specimen PLB-0, PLB-3, PLB-5 and PLB-7.

Based on the Table 1, it is seen that the mechanical properties of ETP/PE polyblend increase with increasing amount of LNR addition. The specimen with 5% weight of LNR addition (PLB-5) shows yield and tensile strength values of 106 $Kg.cm^{-2}$ and 91 $Kg.cm^{-2}$ respectively. This means that the PLB-5 specimen has most rigid specimen compared to the othes. It is shown that high tensile and yield strength values of the PLB-5 specimen represent the optimum variation composition of ETP/PE/LNR polyblend composite materials. To determine visually the surface shape of tested materials, further specimens were observed by SEM, as shown in Figure 3.

Microstructure

The microstructure of pure polyblend without LNR content (PLB-0) is shown in Figure 3(a), while microstructure of polyblend with LNR content of 3% (PLB-3), 5% (PLB-5) and 7% (PLB-7) by weight were mentioned in Figures 3(b), 3(c) and 3(d), respectively.

Table 1. Result of density test

No	Specimens	Density ($g.cm^{-3}$)
4	PLB-0	0.4023
5	PLB-3	0.4785
6	PLB-5	0.5189
7	PLB-7	0.7546

Table 2. Result of mechanical test

Specimen	Yield	Tensile	EB (%)
	Strength ($Kg.cm^{-2}$)		
PLB-0	61	56	164
PLB-3	102	182	660
PLB-5	106	191	650
PLB-7	105	181	600

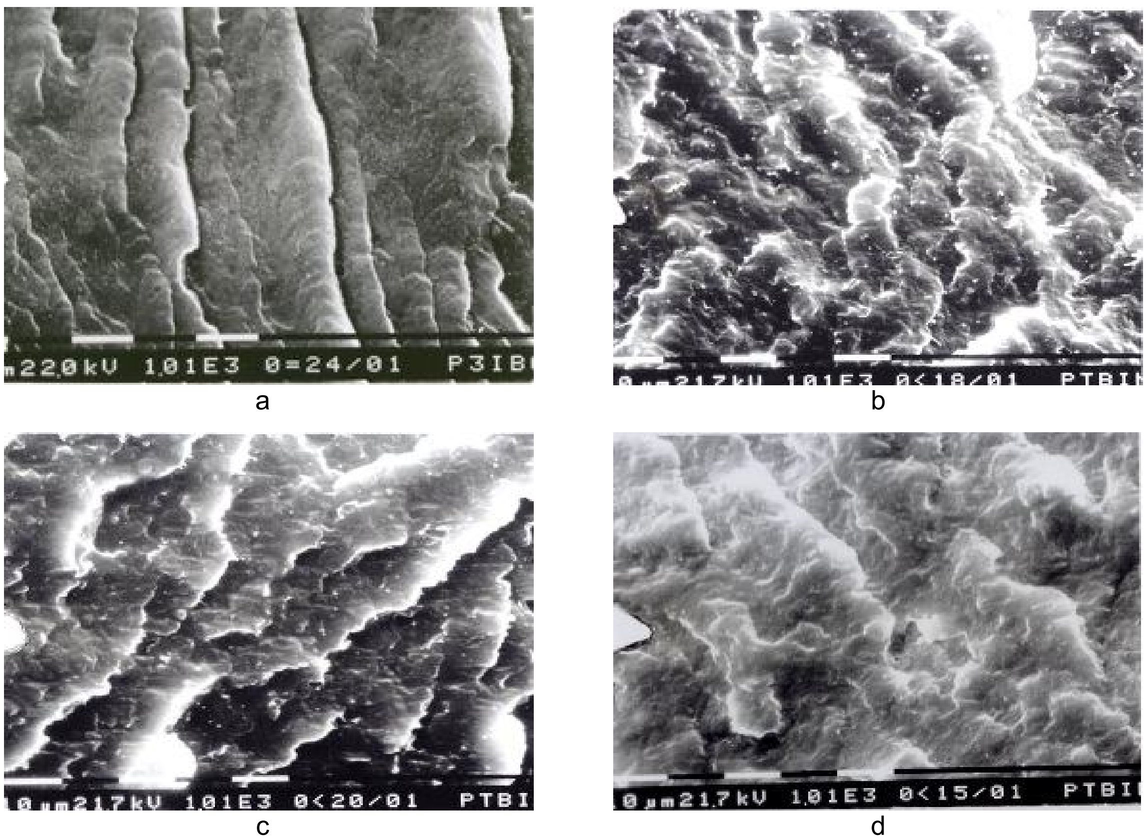

Figure. 3. SEM micrograph of *poly-blend* ETP/PE with variation composition of LNR from specimen (a). PLB-0, (b). PLB-3, (c). PLB-5 and (d). PLB-7.

According to Figure 3, it is demonstrated that the specimen PLB- 0 (Figure 3(a)) has very low mechanical property, see Table 2. The microstructure changed with addition of LNR to the matrix PE/TPE polyblend, Figures 3(b), 3(c) and 3(d). It means that the materials formed PE/ETP polyblend–LNR composite materials as called PLB. It is obvious that the specimen of PLB-3 and PLB-7 (Figures 3(b) and 3(d)) have irregular and abundant microstructure, where as for specimen of PLB-5 shows more regular and homogeneous microstructure (Figure 3(c)). This condition represents that addition of 5% weight of LNR has optimum composition of ETP/PE polyblend–LNR composite materials.

It can be concluded that mixing between ETP/PE polyblend and 5% weight of LNR as compatibilizer may form homogenous and high grain ordered composite materials having high mechanical properties.

CONCLUSION

Based on the result of this research, it can be concluded that the process of TPE/PE polyblend with mixture of LNR 0%, 3%, 5% and 7% by weight have succeeded to be conducted. The mixing materials may form TPE/PE polyblend–LNR composite materials. The density of TPE/PE polyblend–LNR composite materials shows higher density and melting point then the TPE/PE poly-blend origin materials, but the decomposition temperature decreased. The mechanical properties of TPE/PE polyblend origin materials increased with increasing filler LNR for all variation composition. The highest mechanical property is reached for 5% weight of LNR filler content. This condition represents the optimum composition of ETP/PE/LNR polyblend showing regular and homogenous microstructure.

ACKNOWLEDGEMENT

The author would like to thank to the Head of Center for Technology of Nuclear Industry materials. That way also thanks to Mrs. Anik S who assist for blending and mechanical test. This work has been supported by Blok Grant Research 2009.

REFERENCES

Dahlan, H.M, M.D Khairul Zaman A, and A Ibrahim.2002. *Liquid Natural Rubber (LNR) as a Compatibilser in NR/LLDPE Blends—II: The Effects of Electron Beam (EB) Irradiation, Radiation Physics and Chemistry*, Volume 64, Issues 5-6: 429-436.

Deswita, Aloma Karo karo, Sudirman, and Indra Gunawan. 2008. Modifikasi Polietilen Sebagai Polimer Biodegradable Untuk Bahan Kemasan, *Indonesian Journal of Science Materials*, Special Edition, 43-47.

Deswita, Aloma Karo karo, Sudirman, and Indra Gunawan.2002. Pengaruh Penambahan Filter Jerami Terhadap Sifat Mekanik dan Termal Komposit Berbasis Polipropilene, Prosiding Pertemuan Ilmiah IPTEK Bahan *'02, P3IB-BATAN*, Serpong: 225-228.

Deswita, Sudirman, Aloma Karo karo, and Indra Gunawan. 2008. Pengaruh Penambahan Liquid Natural Rubber Pada Polyblend Elastomer Termoplastik-Polipropilen, *Jurnal Sains Materi Indonesia*, Vol.9 No.2, ISSN 1411-1098: 156-160.

Deswita, Sudirman, Aloma Karo karo, Sugik Sugiantoro, and Ari Handayani. 2006. Pengembangan Elastomer Thermo-plastik Berbasis Karet Alam dengan Polietilen dan Polipropilen untuk Bahan Industri*, Indonesian Journal of Materials Science*, *Vol.8 No.2: 52-57.*

Envis. *Indian Center for Plastic in Environmental, India*, 2003.

Fink, J.K. 2010. "Handbook of Engineering and Specialty Thermoplastic: Polyolefins and styrenics." *Scrivener Publishing*: 35.

Halimatudahliana, Nasir M, and Ismail H. *2001. Compatibilizing Effect of Ionomer on Mechanical Properties of Polystyrene and Polypropylene Blend, National Symposium on Polymeric Material 2000, USM: 211-215*

Ismail, H, and Suryadiansyah. 2002. Thermoplastic Elastomeric Based On Polypropylene/Natural Rubber and Polypropylene/Recycle Rubber Blends, *Polymer Testing*, Volume 21, Issue 4: 389-395.

Mahsuri, Ana Marini, and Sudirman. 2005. *Pengaruh Anti Oksidan terhadap Kestabilan Sifat Fisis Bahan Polipaduan Polipropilena-Karet Alam: I. Studi Morfologi dan Sifat Mekanik*, Jurnal Fisika and Aplikasinya, Department Physical, FMIPA-ITS, Volume 1, Nomor 2:050203-1-050203-5

Mashuri, Kristiawan Setia H, Darminto, Aloma K.K, and Sudirman. Pengaruh Irganok 245 Terhadap Kristalinitas dan Kestabilan Mekanik Polipaduan Polipropilen-Natural Rubber, *Jurnal*

Sains Materi Indonesia, Vol.9 No.3 Juni 2008, ISSN 1411-1098, hal. 208-213.

Olk, Olaf Pena, and Ralf H. 2004. *Method For Anchoring an Elastomeric Into a Thermoplastic During Injection Molding, US Patent 20040222254,*

Osha. *Polymer Matrix Materials: Advanced C-32omposites*. 2010: 29-32.

Sudirman, Ari Handayani, Tri Darwinto, Teguh YSP, Anik Sunarni, and Isni Marlijanti. 2000. Struktur mikro dan Sifat Mekanik Komposit Elastomer Termoplastik-Timbal Oksida*, J. Mikroskopi dan Mikroanalisis, Vol 3 No 1: 17-20.*

Utama, Marga, Kadarijah, Herwinarni, Made Sumarti, and F.X Marsongko. *1995*. *Pembuatan Elastomer Termoplastik Karet Alam dengan Metode Polimerisasi Iradiasi*, National Symposium – Indonesian Polymer Association, Jakarta.