[Makara Journal of Technology](https://scholarhub.ui.ac.id/mjt)

[Volume 17](https://scholarhub.ui.ac.id/mjt/vol17) | [Issue 3](https://scholarhub.ui.ac.id/mjt/vol17/iss3) Article 3

8-3-2013

The Effect Zeolite Addition in Natural Rubber Polypropylene Composite on Mechanical, Structure, and Thermal Characteristics

Nurdin Bukit

Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Negeri Medan, Medan 20221, Indonesia, nurdinbukit5@gmail.com

Erna Frida Department of Engineering, University Quality of Medan, Medan 20221, Indonesia

Follow this and additional works at: [https://scholarhub.ui.ac.id/mjt](https://scholarhub.ui.ac.id/mjt?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages)

Part of the [Chemical Engineering Commons](https://network.bepress.com/hgg/discipline/240?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages), [Civil Engineering Commons,](https://network.bepress.com/hgg/discipline/252?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages) [Computer Engineering](https://network.bepress.com/hgg/discipline/258?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages) [Commons](https://network.bepress.com/hgg/discipline/258?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages), [Electrical and Electronics Commons,](https://network.bepress.com/hgg/discipline/270?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages) [Metallurgy Commons,](https://network.bepress.com/hgg/discipline/288?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages) [Ocean Engineering Commons,](https://network.bepress.com/hgg/discipline/302?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages) and the [Structural Engineering Commons](https://network.bepress.com/hgg/discipline/256?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages)

Recommended Citation

Bukit, Nurdin and Frida, Erna (2013) "The Effect Zeolite Addition in Natural Rubber Polypropylene Composite on Mechanical, Structure, and Thermal Characteristics," Makara Journal of Technology: Vol. 17: Iss. 3, Article 3. DOI: 10.7454/mst.v17i3.2926 Available at: [https://scholarhub.ui.ac.id/mjt/vol17/iss3/3](https://scholarhub.ui.ac.id/mjt/vol17/iss3/3?utm_source=scholarhub.ui.ac.id%2Fmjt%2Fvol17%2Fiss3%2F3&utm_medium=PDF&utm_campaign=PDFCoverPages)

This Article is brought to you for free and open access by the Universitas Indonesia at UI Scholars Hub. It has been accepted for inclusion in Makara Journal of Technology by an authorized editor of UI Scholars Hub.

The Effect Zeolite Addition in Natural Rubber Polypropylene Composite on Mechanical, Structure, and Thermal Characteristics

Nurdin Bukit^{1*} and Erna Frida²

1. Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Negeri Medan, Medan 20221, Indonesia 2. Department of Engineering, University Quality of Medan, Medan 20221, Indonesia

** e-mail: nurdinbukit5@gmail.com*

Abstract

This study was conducted to determine the effect of zeolite as filler on mechanical properties, and thermal structure blends of natural rubber and polypropylene (NR/PP). In this study, NR/PP/PP-g-MA blends was filled uncalcined zeolite and calcined zeolite at different weight percent of the 2, 4, and 6%. Samples were characterized to determine the tensile strength, fracture elongation and Young's modulus. Structure of the samples was investigated using XRD method and thermal characteristics were analyzed using DTA/TGA technique. The results obtained for tensile strength showed a significant influence with the addition zeolite of the NR/PP blends at 2% by weight of the composition (8 MPa) and decreased with increasing zeolite composition uncalcined zeolite or calcined zeolite. At 2% to 6% by weight of an increase in Young's modulus, for which uncalcined zeolite function more effectively than the calcined zeolite. On the other hand, the addition of uncalcined zeolite and calcined zeolite did not result in decreased fracture elongation. The XRD analysis zeolite indicate the existence of amorphous phase and crystalline mineral type mordenite $(CaAl₂Si₁₀O₂₄·7 H₂O)$. In general, cristality of samples filled with calcined zeolite slightly higher than the uncalcined zeolite at the same amount and intercalation occurs between the zeolite matrix NR/PP. Thermal analysis showed that the samples are filled with calcined zeolite has a thermal resistance higher than the uncalcined zeolite, the melting point NR/PP 165.0 °C with the addition of 6 wt% zeolite melting point to 186.7 °C.

Abstrak

Pengaruh Penambahan Zeolit pada Komposit Karet Alam Polipropilena terhadap Sifat Mekanik, Struktur dan Termal. Penelitian ini dilakukan untuk mengetahui pengaruh zeolit sebagai bahan pengisi terhadap sifat mekanik, struktur dan termal dari campuran karet alam dan polypropylene (NR/PP). Dalam penelitian ini, campuran NR/PP/PP-g-MA diisi dengan zeolit tanpa kalsinasi dan zeolit kalsinasi pada persen berat yang berbeda yakni 2, 4, dan 6%. Sampel dikarakterisasi untuk menentukan kekuatan tarik, perpanjangan putus dan modulus Young. Struktur sampel diteliti menggunakan metode XRD dan karakteristik termal dianalisis dengan menggunakan DTA/TGA. Hasil yang diperoleh untuk kekuatan tarik menunjukkan adanya pengaruh yang signifikan dengan penambahan zeolit pada campuran NR/PP pada komposisi 2% berat (8 MPa) dan terjadi penurunan dengan bertambahnya komposisi zeolit baik kalsinasi maupun tanpa kalsinasi. Pada 2% sampai 6% berat modulus Young terjadi peningkatan, untuk zeolit yang tidak dikalsinasi berfungsi lebih efektif dibandingkan dengan zeolit kalsinasi. Di sisi lain, penambahan zeolit kalsinasi dan tidak dikalsinasi menghasilkan penurunan perpanjangan putus. Analisis XRD dari zeolit menunjukkan adanya fase amorf dan kristal tipe mineral mordenite (CaAl₂Si₁₀O₂₄.7 H₂O). Secara umum, kristalitas dari sampel yang diisi dengan zeolit kalsinasi sedikit lebih tinggi dari pada zeolit tanpa kalsinasi pada jumlah yang sama dan terjadi interkalasi antara zeolit dengan matrik NR/PP. Analisis termal menunjukan bahwa sampel yang diisi dengan zeolit kalsinasi memiliki daya tahan termal yang lebih tinggi dibandingkan dengan zeolit tanpa kalsinasi, titik leleh NR/PP 165,0 °C dengan penambahan zeolit 6% berat titik leleh menjadi 186,7 °C.

Keywords: mechanical, natural rubber, structure, thermal, zeolite

1. Introduction

Natural rubber has been known as raw material for many various applications, such as rubber belt, fan belt, automotive brake pad, tyres, and many others. As an important type of polymer material, rubber is widely used due to its high and reversible deformability. However, since the essential modulus and mechanical

strength as well as thermal resistance are low, natural rubber is commonly blended with other materials, one of them is polypropylene (PP). Several studies have reported that blending of natural rubber with PP resulted in characteristic improvement, such as increased thermal stability [1], decreased fracture elongation [2], and increased Young's modulus [3]. However, the NR/PP was found to exhibit low abrasion resistance, tear strength, and high rolling strength, which make it unsuitable for many practical uses of rubber materials, such as tire.

In polymer science, it has been acknowledged that the properties of polymer materials could be strengthen by addition of different kinds of inorganic fillers. Furthermore, it has been found that the size and dispersion characteristics of inorganic particles have a great effect on the comprehensive properties of polymer composites [4-6]. These findings reflect that the application of nano-size fillers is a promising method for property modification of polymer materials. To take advantage of the natures of filler, many attempts have been made to improve the properties of NR/PP blend by utilization of various inorganic fillers, such as silica [7- 8], carbon [9], zeolite [10], and bentonite [11]. Addition of filler is aimed not only to fill a rubber matrix with small particles, but also to modify the matrix texture as a result of interaction of rubber molecules with the filler particles during processing. By addition of proper filler, it is then possible to produce modified materials with improved properties, such as thermal and mechanical properties. As an example, filler with high specific surface area is expected to enhance interphase effects and tensile strength of the rubber blend [12].

The effect of filler depends essentially on the efficiency of the dispersion of the fillers and the interphase between the rubber molecules and the particles of the filler. In many cases, the dispersion of filler particles into rubber moieties is problematic due to possible entanglement of the particles and high viscosity of rubber. For this reason, blending of rubber with compatibilizer, such as polypropylene grafted maleic anhydride (PP-g-MA) [13] prior to addition of filler is commonly practiced. Addition of compatibilizer is required in order to improve the homogeneity of the blend, therefore, facilitating the distribution of the filler particles.

This study preparation made purification of natural zeolite with HCl solution and then calcined the preparation of nano size particles made of natural zeolite with a ball mill process. Result of natural zeolite nano particles are used as fillers in mixed NR/PP was carried out to investigate the effect of nano zeolite addition on thermal and mechanical NR/PP blend properties with PP-g-MA compatibilizer. Nano zeolite was used because this mineral is stable at hight temperatures, resistant to organic solvents, chemicals, and has high tensile strength as well as hardness. Structurally, zeolite is known to have very reguar crystalline form with cavities that are interconnected in all directions, making it very resistant to high thermal treatment [14-15]. Based on these properties, zeolite is considered as good reinforcing agent for improving mechanical and thermal properties organic polymers. To take advantage of its reinforcing capacity, utilization of zeolite as filler for different types of polymer materials has been discussed in many studies. In particular it is reported that utilization of zeolite led to significant increase of tensile strength and thermal resistance of natural rubber composite [12,16]. In recognition of these properties, it is then expected that addition of zeolite will significantly improve the mechanical and thermal properties of the NR/PP blend.

A part from its potential as reinforcing agent, natural zeolites are known as hydrophilic materials, making them less compatible with most organic polymers which are known to posses hydrophobic surface. For this reason, modification of zeolite to increase its surface hydrophobicity is required to promote the compatibility of zeolite and organic polymers. The most common modification methods applied are thermal treatment at temperature of $600-1100$ °C $[17-19]$ and chemical modification using strong acids or strong bases [20-21]. In this study, natural zeolite was modified by calcination at 600 °C to activate the zeolite while retaining its amorphous state [17]*.*

Polymer nano composites are two-phase materials in which the polymers are reinforced by nano scale fillers. The most heavily used filler material is based on the smectite class of aluminum silicate clays, of which the most common representative is montmorillonite (MMT). MMT has been employed in many polymer layered silicate (PLS) nano composite systems because it has a potentially high-aspect ratio and high-surface area that could lead to materials which could possibly exhibit great property enhancements. In addition, it is environmentally friendly, naturally occurring, and readily available in large quantities. Layered silicates in their pristine state are hydrophilic. Most of the engineering polymers are hydrophobic [22].

2. Methods

Materials. The materials used in this study include, natural rubber SIR-20, from PT Santos Ruber Jakarta, polypropylene copolymer was obtained from Production Singapore. This polymer has melting point of 176 °C, and density of 0.896 g/cm³. Polypropylene grafted maleic anhydride (PP-g-MA) was obtained from Japan. Zeolite were obtained from Pahae, North Tapanuli District of North Sumatra Province. Carbon Black N 330 was obtained from Cilegon Indonesia. Cabot Red Seal Zinc oxide was obtained from Aldrich. Sulphur and

N-Isopropyl-N-Phenyl-p-phenylenediamine (IPPD) were obtained from Qingdao China. Tetra methyl thiurea disulfarat (TMTD), Marcapto benzhoathizole disulfide (MBTS), and stearic acid were purchased from Sumi Asih Indonesia.

Instrumentation. Two roll mixing mill of Internal Mixer Laboplastomil Model 30 R150, X-ray difractometer type PW 1710 with voltage of 40 kV and current of 25mA, Hydraulic hot press, cold press of 37 ton from Genno Japan, Ball mill HEM–E3D, Planetary ball mill PBM-4, Particle Size Analyzer DeslaTM nano, Universal Testing Mechanic model Laryee Universal Testing Machine Wdw-10, for simultaneous thermal analysis (DTA-TGA) Setaram TAG24.

Preparation of nano zeolite. In this study, both uncalcined and calcined zeolite was used as filler. Uncalcined nano-zeolite was prepared by grinding the chunks of zeolite to obtain the powder with the size of 200 mesh, the sample was grinded using planetary ball mill PBM 4 for 10 hours the zeolite was found to have the particle size of 144.8 nm. Preparation of calcined zeolite was carried in similar way, but the powder was the size of 200 mesh purified using 2 M HCl solution with the ratio of zeolite (mass) to HCl solution (mL) of 1:10, by stirring the mixture using magnetic stirer for 2 hours. The sample was subjected to calcination treatment at 600 °C for 2 hours, and then grinded for 10 hours using planetary ball mill PBM 4. After this treatment, the calcined zeolite was found to have the particle size of 190.2 nm [18]. Analyses XRF in Table 3 XRD analysis of natural zeolite obtained by purification and calcination phase containing major mineral type mordenite (CaAl₂Si₁₀O₂₄.7 H₂O).

Preparation of natural rubber compound. Natural rubber compound was prepared by mixing natural rubber (SIR-20) was mixed with blended using two roll mixing mill with the formula as shown in Table 1. Preparation of the sample was commenced by grinding natural rubber for five minutes, to transform it into crumb, followed by addition of wax and carbon black. While the sample was continuously grinded for another five minutes, zinc oxide, stearic acid, IPPD, and sulphur

were added into the mixture, followed by introduction of TMTD after the process last for 13 minutes and MBTS after 18 minutes. The process was completed after the whole mixture was grinded for five minutes. The compound obtained from this treatment was then used to prepare NR/PP blend loaded with zeolite as filler.

Preparation of compound NR/PP blend. To prepare coumpound NR/PP blend, the natural rubber compound previously prepared was mixed with PP, PP-g-MA, and zeolite, according to the composition as presented in Table 2. The mixture was blended using Laboplastomil Model 30 R150 internal mixer with a volume of 60 cc. The blending was carried out at temperature of 178 °C with rotor speed of 60 rpm for 10 minutes.

Mechanical properties measurement. Tensile strength measurement was performed according to JIS K 6781 standard using a Laryee Universal Testing Machine Wdw-10, at crosshead speed of 50 mm min⁻¹. Young's modulus (E), ultimate tensile strength (σ_{max}), and fracture elogation (ε_b) were determined from the stressstrain curves.

XRD analysis. The XRD analysis was conducted at room temperature using X-ray difractometer type PW 1710. The operating conditions used were CuK₁ radiation (λ = 0.15418 Å, produced at 40 kV and 25mA). Pattern was recorded over goniometer (2θ) ranging from 5° to 65°.

Table 1. The Formula Used for Preparation of Natural Rubber Compound

Ingredients	Quantity (phr	Function
$SIR-20$	100	Binder
Wax	1.5	Antilux
Carbon BlackN 330	30	filler
Zinc oxide	5	Activator
Stearic acid	\mathcal{L}	Activator
Sulfur	3	Curing agent
IPPD	\mathfrak{D}	Antioxidant
TMTD	1.5	Accelerator
MBTS	2.5	Accelerator

XRF				
Calcined Zeolite		Uncalcined Zeolite		
Chemical	Composition	Chemical	Composition	
Composition	$(wt\%)$	Composition	$(wt\%)$	
Na ₂ O	1.03	Na ₂ O	1.76	
MgO	0.82	MgO	0.12	
Al_2O_3	17.29	Al_2O_3	14.19	
SiO ₂	71.06	SiO ₂	80.30	

Table 3. Chemical Composition Nano Particle Zeolite of XRF

The interlayer distance of zeolite in nanocomposite was derived from the peak position $(d_{001} =$ reflection) in XRD diffractograms according to the Bragg equation: *d* $=\lambda$ /2sin θ .

 K_2O 4.63 K_2O 1.45 CaO 1.92 CaO 0.14 TiO_2 0.51 TiO_2 0.52 $Fe₂O₃$ 2.75 Fe₂O₃ 0.91

 P_2O_5 0.61

Thermal analysis. Simultaneous thermal analysis (STA) was performed on TGA-DTA Setaram TAG 24, by scanning the sample over the temperature range of 50–600 °C using nitrogen gas with the flow rate of 50 mL/min and a heating rate of 20 °C/min.

3. Results and Discussion

Mechanical characteristics. In this study, mechanical properties of the samples include tensile strength, Fracture elongation, and Young's modulus, are measured in order to evaluate the effect of the quantity of zeolite used and to compare the performance of uncalcined and calcined zeolite in Table 4. Figure 1 shows the tensile strengths of the samples filled with zeolite at different quantities (wt%).

As can be seen in Figure 1, the samples filled with zeolite display higher tensile strength compared to the sample without zeolite, implying that adition of zeolite led to increased ability of the samples to withstand breaking pressure. This ability is in accrodance with the nature of zeolite as rigid material, therefore, increased the rigidity of the NR/PP composite. In general, uncalcined and calcined zeolite were found to display similar trend, in which the highest tensile strength was displayed by the sample filled with 2% zeolite, and decreased with increased quantities of the zeolite used. This result appears to suggest that at small amount, the zeolite particles interact homogeneously with the rubber matrix to produce effective interphase, which function to redeem the pressure acting on the composite. At higher zeolite quantities, decreased strength observed is most likley caused by the formation of voids around the filler due to poor bonding between zeolite and the rubber matrix, as suggested by other workers [23]. As

shown by Figure 1, the patterns observed for the samples filled with uncalcined and calcined zeolite are practically similar. However, the tensile sterength of the sample filled with uncalcined zeolite is relatively lower than that of the sample filled with calcined zeolite at the same amount. Better performance of calcined zeolite is most likely the contribution of two factors, namely higher hidrophobicity due to acid and thremal treatments and smaller particle size [24].

Fracture elongation of the samples filled with uncalcined and calcined zeolite are shown in Figure 2, indicating that uncalcined and calcined zeolite imparted quite different effects on the NR/PP blend.

The results obtained demonstrated that for the samples filled with uncalcined zeolite, the fracture elongation of the sample without zeolite is higher than those of the samples filled with zeolite. It is also found that increased amount of zeolite led to decreased fracture elongation, which is in accordance with the nature of silica which is known to have very small elasticity. This change in fracture elongation is most likely attributed to the interaction of the zeolite with natural rubber moiety. During the blending, the particles of zeolite were scattered randomly, producing larger surface contact area which result in strong binding of the zeolite particles with the NR/PP blend.

For the samples filled with calcined zeolite, it can be seen that the fracture elongation of the samples filled with 2 and 4% zeolite are higher than that of the sample without filler, while for the sample filled with 6% zeolite, the opposite is true. As can be seen, the maximum value of fracture elongation was displayed by the sample filled with 2% zeolite and continue to decrease with an increase of filler content. This reduction of fracture elongation is most likely due to reduction of in the effective matrix cross-section and formation of more voids in the matrix [25].

Young's modulus of the samples are shown in Figure 3, showing the trend of increased modulus with an increase of filler content. In general, the results obtained demonstrated that the samples filled with uncalcined zeolite display higher Young's modulus than that of the sample filled with calcined zeolite at the same amount. The main factor responsible for this difference is probably the smaller particle size of the calcined zeolite, which make the particles easier to penetrate the composite matrix. This trend is in agreement with the trends observed for tensile strength and fracture elongation, since Young's modulus is linearly proportional with tensile strength, while with fracture elongation the opposite is true. Addition of more zeolite will promote the capability of the blend to carry more tensile load as a result of increased tensile strength and decreased fracture elongation. Calcined zeolite tends to

absorb lower hydrocarbon groups (non-polar) natural zeolite activation results tend to absorb hydrocarbon groups are (medium polar) so that the presence of an organic matter content (monmorillonate) in size nono size as the filler material is added to natural rubber [26]. When layered silicates are uniformly dispersed (exfoliated) in a polymer matrix, the composite properties can be improved to a dramatic extent. These improvements may include increased strength, higher modulus [22].

Table 4. Propeties Mechanical Coumpound NR/PP Blends Calcined Zeolite and Uncalcined Zeolite

Zeolite $(wt\%)$	Calcined Uncalcined Zeolite $(wt\%)$	Tensile (MPa)	Fracture Strength Elogation (%)	Young'S Modulus (MPa)
θ	0	6.00	66.90	8.90
\mathfrak{D}	0	8.00	82.83	9.65
4	Ω	6.70	71.25	10.50
6	0	6.30	53.83	11.60
0	\mathfrak{D}	7.70	58.60	13.27
θ	4	6.40	40.16	16.00
	6	5.70	35.83	16.28

Figure 1. Tensile Strength of the Samples Filled with □ Uncalcined and **■** Calcined Zeolite

Figure 2. Fracture Elongation of the Samples Filled with □ Uncalcined and **■ Calcined Zeolite**

Figure 3. Young's Modulus of The Samples Filled with □ Uncalcined and ■ Calcined Zeolite

Figure 4. XRD Patterns of the Samples Filled with Uncalcined Zeolite, (a) 0 wt%, (b) 2 wt%, (c) 4 wt%, and (d) 6 wt%

Figure 5. XRD Patterns of the Samples Filled with Calcined Zeolite, (a) 0 wt%, (b) 2 wt%, (c) 4 wt%, and (d) 6 wt%

X-ray diffraction pattern. The XRD patterns of the samples are presented in Figure 4, displaying the existence of amorphous phase and several crystalline phases. The amorphous phase is most likely associated

with rubber as has been reported previously [27], while the crystalline phases are associated with the zeolite components [28]. The XRD patterns of the samples reinforced with uncalcined zeolite are presented in Figure 4 and the patterns for the samples reinforced with calcined zeolite are presented in Figure 5. In general, it can be seen that no significant difference in term of the phase between the unfilled sample and the samples filled with uncalcined and calcined zeolite. However, it can be observed that the intensities of the peaks in the samples filled with calcined zeolite are relatively higher than those observed for the sample filled with uncalcined zeolite samples, suggesting that the calcined zeolite are more crystalline than uncalcined zeolite. In all samples, it can be seen that the peak with the highest intensity is located at 2θ around 16.8 or d = 5.2730 Å, which is commonly assigned to zeolite [29]. This is supported by the peak at 2 θ of 27.6 or 3.2190 Å, assigned to alumina [30], and that at 2 θ of 23.56 or d = 3.7731 Å, assigned to silica [31-33]. Higher intensities of the peaks associated with zeolite in the samples filled with calcined zeolite are in accordance with the effect of thermal treatment, which is known to convert amorphous materials into crystalline phase as consequence of molecular diffusion during thermal treatment [34]. This higher crystallinity is probably the reason for relatively better mechanical properties of the samples filled with calcined zeolite. Natural rubber mixture with clay nono composite produce significant improvements to the basal spasing of the polymer matrix and show interclation between polymer materials with fillers [35].

Thermal Analysis. In this study, thermal characteristics of the samples were evaluated by analyzing the samples with differential thermal analysis (DTA) and thermal gravimetric analysis (TGA). The DTA thermograms of the samples as filler with different loads of uncalcined zeolite and Calcined Zeolite are compiled in Figure 6 and the corresponding TGA results are presented in Figure 7. The TGA results presented in Figure 7 indicate the existence of six temperature zones, indicating the pattern of weight loss of the samples. At temperature range up to 390 °C, for each of the sample, the weight loss is very small, which is in agreement with the TGA results, showing the endothermic peak associated with the removal of water and volatile components of the rubber compound. At temperature ranging from 390 to 496 °C, the results indicate very sharp weight loss. This part of the thermogram indicate very evidently the ingredients decomposition of the rubber compound and crystallization of zeolite which is supported by the existence of endothermic peak at 496 °C in the DTA thermograms Figure 6. Above 496 °C, the samples are practically have reached stable state, since no more weight loss displayed by the samples shown Figure 7. This means that zeolite particles and rubber chain mixed homogeneously to form compact blend.

Figure 6. DTA Thermograms of the Samples Filled with Uncalcined Zeolite, (a) 2 wt%, (b) 4 wt%, (c) 6 wt% and Calcined Zeolite, (d) 2 wt%, (e) 4 wt%, (f) 6 wt%

Figure 7. TGA Thermograms of the Samples Filled with Uncalcined Zeolite, (a) 2 wt%, (b) 4 wt%, (c) 6 wt% and Calcined Zeolite, (d) 2 wt%, (e) 4 wt%, (f) 6 wt%

Comparing the data for the samples filled with calcined zeolite (Table 6) to those for the samples filled with uncalcined zeolite (Table 5) it can be seen that the effect of the amount of zeolite added is practically similar. It is also found that no significant different in the melting point of the samples filled with uncalcined and calcined zeolite at the same quantity. The main difference observed is considerably higher decomposition temperatures of the samples filled with calcined zeolite than those observed for the samples filled with uncalcined zeolite. The main reason for this difference is most likely higher crystallinity of the calcined zeolite as a result of thermal treatment, as supported by the results of the XRD analysis. Higher crystallinity of calcined zeolite promoted thermal stability of the composite, leading to higher decomposition temperature.

119

Table 5. Thermal Characteristics of the Samples Filled with Uncalcined Zeolite

Sample (wt%)	2	4	6
Melting Point $(^{\circ}C)$		165.3-183.9 167.2-185.6 169.0-185.6	
Decomposition $(^{\circ}C)$		328.9-490.4 359.7-496.5 372.3-495.3	
Endothermic Peak (°C)	465.6	479.4	470.8
Mass Variation (mg)	-4.0	-5.6	-6.7
Mass Variation (%)	-27.7	-38.0	44 A

Table 6. Thermal Characteristics of the Samples Filled with Calcined Zeolite

4. Conclusions

The results obtained clearly revealed the significant effect of zeolite on the characteristics of the NR/PP blend. Addition of zeolite was found to increase the Young's modulus of the samples of 8.9 MPa to 11.6 MPa calcined zeolite an uncalcined zeolite 16.8 MPa, with uncalcined zeolite was found to function more effectively. For tensile strength and fracture elongation, addition of uncalcined zeolite was found to result in decreased values. For these two characteristics, the samples filled with calcined zeolite display higher values than exhibit by the samples filled with uncalcined zeolite at the same quantity. The XRD analysis zeolite indicate the existence of amorphous phase and crystalline mineral type mordenite $(CaAl₂Si₁₀O₂₄·7 H₂O)$ in both uncalcined and calcined of samples. In general, the crytallinity of the sample filled with calcined zeolite is slightly higher than that of the sample filled with uncalcined zeolite at the same quantity and intercalation occurs between the zeolite matrix NR/PP. Thermal analysis showed that the samples are filled with calcined zeolite has a thermal resistance higher than the uncalcined zeolite, the melting point NR/PP 165.0 °C with the addition of 6 wt% zeolite melting point to 186.7 °C.

References

- [1] Bahruddin, Reaktor. 11 (2007) 71.
- [2] E. Frida, Makara Series Technol. 16 (2012) 171.
- [3] W. Xu, G. Liang, H. Zhai, S. Tang, G. Hang, W.P. Pan, J. Eur. Polym. 39 (2003) 1467.
- [4] M.A. Osman, A. Atallah, M. Muller, U.W. Suter. J. Polym. 42 (2001) 6545.
- [5] C.A. Cooper, D. Ravich, D. Lips, J. Mayer, H.D. Wagner, J. Compos. Sci. Technol. 62 (2002) 1105.
- [6] J.S. Kim, D.H. Reneker, Polym. Compos. 20 (1999) 124.
- [7] J.W. Brinketen, S.C. Debnath, L.A.E. M. Reuvenkamp, J.W.M. Noordemeer, Com. Sci. Tech. 68 (2003) 1165.
- [8] S. Siriwardena, H. Ismail, U.S. Ishiaku, J. Polym. Int. 50 (2001) 707.
- [9] L.M. Calvo, S. Noskin, S. Kahan, United States Paten No. #6036998, March 14th, 2000.
- [10] S. Zhang, Y. Li, S. Qu, W. Chen, J. Tribol. Lett. 38 (2010) 135.
- [11] Z. Gu, G. Song, W. Liu, P. Li, L. Gao, H. Li, X. Hu, J. Appl. Clay. Sci. 46 (2009) 241.
- [12] B. Rattanasupa, W. Keawwattana, Kasetsart, J. Natur. Sci. 41 (2007) 239.
- [13] A.C. Karmakar, J. Mat. Sci. Lett. 16 (1997) 462.
- [14] N.S.M. El-Tayeb, K.W. Liew, J. Wear. 266 (2009) 275.
- [15] A. Keskin, Essa, J. Sci. Res. 6 (2011) 4893.
- [16] L. Bokobza, Compos. Inter. 13 (2006) 345.
- [17] Setiadi, A. Pertiwi, Prosiding Kongres dan Simposium Nasional, MKICS, 2C, Semarang, Indonesia, 2007, p.81. [In Indonesia]
- [18] N. Bukit, Makara Series Technol. 16/2 (2012) 121.
- [19] G.E. Christidis, D. Moraetis, E. Keheyan, L. Akhalbedashvili, N. Kekelidze, Gevorkyan, R. Yeritsyan, H. Sargsyan, Appl. Clay Sci. 24 (2003) 79.
- [20] K. Rangsriwatananon, A. Chaisena, C. Thongkasam, J. Porous Mater. 15 (2008) 499.
- [21] P. Kowalczyk, M. Sprynskyy, A. PTerzyk, M. Lebedynets, J. Namiesnik, B. Buszewsk, J. Coll. Interf. Sci. 297 (2006) 77.
- [22] T.Q. Nguyen, G. Donald. Baird Advances in Polymer Technol. 25/4 (2006) 270.
- [23] H. Ismail, L. Mega, Polym. Plast. Tech. Eng. 40 (2001) 463.
- [24] J.R. Leblance, Prog. Polym. Sci. 27 (2002) 627.
- [25] D. Metin, F. Tihminlioglu, D. Balkose, S. Ulku, Composites. Appl. Sci. Manufac. 35 (2004) 23.
- [26] M. Frounchi, S. Dadbin, Z. Salehpour, M. Nofereti, J. Membrane Sci. 282 (2006)142.
- [27] S.Y. Lee, I.A. Kang, G.H. Doh, W.J. Kim, J.S. Kim, H.G. Yoon, Q. Wu, Polym. Lett. 2/2 (2008) 78.
- [28] M. Mondragon, E.M. Hernandez, J.L. Rivera-Armenta, F.J. Rodríguez-González, J. Carbohydr. Polym. 77 (2009) 80.
- [29] M.E. Davis, J. Meso. Mat. 21 (1989) 21.
- [30] D.R. Treadwell, D.M. Dabbs, I.A. Aksay, J. Chem. Mater. 8 (1989) 2056.
- [31] L. Dewimile, B. Bressojn, L. Bokobza, J. Polym. 46 (2005) 4135.
- [32] B. Chaichua, P. Prasassarakich, S. Poompradub, J. Sol-Gel Sci. Tehnol. 52 (2009) 219.
- [33] I.A. Aksay, D.M. Dabbs, M. Sarikaya, J. Am. Ceram. Soc. 74/ 2 (1991) 2343.
- [34] H. Salmah, H. Ismail, A. Bakar, J. Polym. Plast. Technol. Eng. 44 (2005) 863.
- [35] P.N. Alam, T. Rihayat, Rekayasa Kimia dan Lingkungan. 16 (2007) 1. [In Indonesia]