EEET ECOLOGICAL ENGINEERING & ENVIRONMENTAL TECHNOLOGY

Ecological Engineering & Environmental Technology 2023, 24(2), 205–213
https://doi.org/10.12912/27197050/156961
ISSN 2719-7050, License CC-BY 4.0

Received: 2022.10.09
Accepted: 2022.12.16
Published: 2023.01.01

Effect of Hybrid Filler Oil Palm Boiler Ash – Bentonite on Thermal Characteristics of Natural Rubber Compounds

Erna Frida[1], Nurdin Bukit[2*], Ferry Rahmat Astianta Bukit[3], Bunga Fisikanta Bukit[4]

[1] Department of Physics, Universitas Sumatera Utara, 20155, Medan, Indonesia
[2] Department of Physics, Universitas Negeri Medan, 2022, Medan, Indonesia
[3] Department of Electrical Engineering, Universitas Sumatera Utara, 20155, Medan, Indonesia
[4] Universitas Quality Berastagi, 22153, Berastagi, Indonesia
* Correspouding author's e-mail: nurdinbukit5@gmail.com

ABSTRACT

The concept of hybrid fillers by combining several types of fillers, especially with the inclusion of nanoscale filler particles, has attracted the attention of many researchers. The addition of filler to the rubber compound can improve the thermal properties. This study aims to analyze the effect of filler hybrid oil palm boiler ash (OPBA) –bentonite on thermal characteristics natural rubber compounds. The coprecipitation method was used in preparing OPBA, and CTAB surfactant was added in bentonite preparation. Meanwhile, compound preparation was carried out by inserting SIR 20 into an open mill machine. Characterization was done by XRD, FTIR, SEM, mechanical and thermal properties. In general, the compounds did not show a change in peak position. Peaks of $2\Theta = 44^{o}$ and 64^{o}. Bonds at 2849–2917 cm^{-1} were associated with asymmetric methyl stretching vibrations. The peaks of 1000–650 cm^{-1} showed C = C-H bending. The compound morphology shows torn lines with branching. Furthermore, in general, the mechanical properties of the compound increased with the addition of OPBA/bentonite filler. Differential Scanning Calorimetry compound data showed an increase in the number of peaks in the sample with 10 wt% filler.

Keywords: filler, rubber, compound.

INTRODUCTION

Strengthening with various fillers is very important for rubber, as unfilled rubber has minimal applications due to its poor thermal and physical properties. Rubber with filler material has been widely applied in various industrial fields because of its outstanding mechanical properties, thermal stability, and good oil resistance. Several fillers have been used in rubber manufacturing as reinforcing agents for NR composites, including fly ash, silica nanoparticles, carbon black, palm ash, potato starch, cellulose, and agroindustry residues (Bukit et al., 2022a; Bukit et al., 2022b; Bukit et al., 2019; Farida et al., 2019; Panitchakarn et al., 2019). According to previous occupational theory rubber strengthening by nanoparticles is associated with the formation of stretch polymer chains straight between adjacent particles, caused by slippage of the adsorbed polymer chains on the filler surface during tension. Formation of stretched straight polymer chains depending on filler content, filler dispersion,determined mainly by filler-filler interactions and filler-rubber interactions. Filler content and dispersion determine the distance between adjacent particles, which should be small sufficient to ensure that the cross-linked rubber chains can attached to at least two different particles (chains bridge), and the filler-rubber interaction is critical for slip polymer chains on the surface of the filler particles to form stretch straight polymer chains (Wang et al., 2010).

Carbon black (CB) and precipitated silica is widely used as rubber reinforcing fillers. Incorporating CB or silica during compounding will increase the strength-related properties of the

compound rubber to some extent. However, both fillers have their limitations, which have prompted scientists and researchers to develop new alternative fillers, either to replace or partially replace these conventional fillers and reduce their interdependence. Substances such as organophilic modified clay and bentonite are possible future filler systems (Stöckelhuber et al., 2010).

The concept of hybrid fillers by combining several types of fillers, especially with the inclusion of nanoscale filler particles, has attracted the attention of many researchers. It is revealed that multiphase hybrid fillers can maintain superior properties to all fillers and exhibit synergistic effect processing in polymers and elastomers due to compatibility and cooperative interactions at the nanoscale level (Senthivel et al., 2015)

According to the research, a synergistic effect has been found in the hybrid reinforcement based on precipitated silica and montmorillonite nanofiller. Combining montmorillonite and silica as fillers produces balanced rubber properties (Bao et al., 2015). Ismail and Haw studied the effect of hybrid palm ash/silica filler on rubber. In this study, Rubber compounds were prepared on different OPBA/bentonite hybrid compounds and were referenced to study the effect of filler content on the properties of the rubber compounds. Similar results were obtained when a hybrid slurry/silica paper filler was introduced into the rubber matrix (Abdul Salim et al., 2018; Ismail & Haw, 2010). The study of the thermal transition of polymers is very important in the design of polymer products for know the temperature range of the material in order to used without degradation. The composition of the rubber compound may vary depending on the purpose for which the finished product is made. The stages of the rubber compound manufacturing process include mixing, forming, and vulcanization. This research examines the mechanical and thermal properties of the rubber compound, which is given a filler with various composition variations.

MATERIAL AND METHOD

Material

OPBA from PT. DPI (Dhajaja Putra Indonesia) Asahan District North Sumatra Indonesia, Bentonite, 6M HCL, NH_4OH Merck Pro Analis, CTAB, SIR-20 rubber, wax, ZnO, stearic acid, sulfur, BHT, ZDEC

Method

Preparation OPBA and bentonite nanoparticle

OPBA waste was dried and calcined at 500°C for 5 hours and then milled with a ball mill type Planetary Ball Mill for 10 hours with a rotation of 250 rpm filtered using a 200 mesh sieve. Then, Mixed OPBA with 6M HCl at 70°C for 4 hours. OPBA was mixed with NH_4OH at 70°C for 4 hours and then neutralizing the pH. Meanwhile, bentonite was calcined for 5 hours at 700°C. Milling bentonite with a ball mill for 10 hours with a rotation of 250 rpm. 0.2 moles of CTAB are mixed with distilled water. Bentonite, CTAB, and distilled water was mixed at a temperature of 100°C for 4 hours (Frida et al., 2022).

Preparation of rubber compound with OPBA and bentonite filler

Compound preparation was carried out by inserting SIR 20 into an open mill machine. Then, put the compound into a standard slab mold container. Then, cut the slab using a shaper that conforms to the Dumb-Bell test standard into a specimen shape. The composition of the compound with OPBA filler and bentonite in phr is shown in Table 1. Meanwhile the compound preparation process is shown in Table 2.

RESULTS AND DISCUSSION

XRD analysis of rubber compound

XRD characterization is useful for obtaining diffraction patterns and crystal structures. The XRD used is the Shimadzu 6100 type (40 kV, 30 mA) with a wavelength of Cu-Kα1 = 1.5405 = 0.15406 nm, with a rate of 2°/min at an angle range of 2Θ = 5°–70°. Figure 1 shows XRD of compound with OPBA and bentonite filler.

In general, the compound showed no change in the peak position. Peak 2Θ = 44°, 64° and dhkl {-130,22-2} are exist in all samples. An increase in the amount of filler indicates an increase in intensity. However, the compound containing filler (4, 6, 10) wt% showed a decrease in intensity. Moreover, higher filler loading revealed higher peak intensity values. It is due to the agglomeration of Bentonite and silicate layer OPBA at higher filler loads. To explain the results, we must consider the following three factors: (1) group size, (2) polymer polarity, and (3) chain

Table 1. The composition of the compound with OPBA filler and bentonite

No	Materials	Compound formula (perhundred rubber)						Function
		S_0	S_1	S_2	S_3	S_4	S_5	
1.	SIR-20 rubber	92	92	92	92	92	92	Binder
2.	OPBA/ Bentonite nanoparticle	0	2	4	6	8	10	Filler
3.	Wax	1.5	1.5	1.5	1.5	1.5	1.5	Antilux
4.	ZnO	5	5	5	5	5	5	Activator
5.	Stearic acid	2	2	2	2	2	2	Activator
6.	Sulfur	3	3	3	3	3	3	Curing agent
7.	BHT	2	2	2	2	2	2	Antioksidan
8.	ZDEC	4	4	4	4	4	4	Accelerator

Table 2. The compound preparation process

No	Mixing steps	Time (minutes)					
		S0*	S1	S2	S3	S4	S5
1	Rubber mastication	3	3	3	3	3	3
2	Addition of ZnO and stearic acid	2	2	2	2	2	2
3	Added PPD and filler	1	1	1	1	1	1
4	Addition of TMTD and MBTS	1	1	1	1	1	1
5	Addition of sulfur	1	1	1	1	1	1
6	Blending	1	1	1	1	1	1
7	Remilling	1	1	1	1	1	1
Total time		10	10	10	10	10	10

* S0 without filler.

segmental mobility, as the structure of the clay is the same. Also, the polymer chain lengths are assumed to be similar since the molecular weights of these polymers are approximate of the same order (Sadhu & Bhowmick, 2004).

FTIR analysis of rubber compound

FTIR analysis was carried out to determine changes in functional groups that occurred in the compound. Changes in functional groups experienced by the compound indicate a chemical interaction between Natural rubber and filler. This infrared spectrum is analyzed by observing the typical frequencies of the functional group of the sample FTIR spectra. The FTIR used is the Agilent Cary 630 FTIR. This flexible benchtop FTIR instrument that offers high performance and exceptional ease of use in an ultra-compact design.

FTIR characterization of rubber compound with OPBA/bentonite filler is shown in Figure 2. Bonding in the 2849–2917 cm^{-1} is associated with asymmetric methyl stretching vibrations, methyl asymmetric deformation vibrations, and methyl symmetric deformation (Xu et al., 2015). Peak 1000–650 cm^{-1} indicates C = C-H bending. The peaks of 1514–1391 cm^{-1} also show C-H bending. The FTIR spectrum between natural rubber compounds with the addition of OPBA and bentonite fillers and without fillers did not show a significant difference. This was probably because the fillers were evenly distributed in the rubber compounds and maybe also because of the amount of filler that did not significantly affect the bond in natural rubber (Farida et al., 2019; Ginting et al., 2020).

SEM analysis of rubber compound

scanning electron Microscope can provide information about the surface topography of a specimen. SEM characterization was carried out using the SEM TM3030 model. Hitachi High-Tech has provided a “5 kV mode” that allows for sharper observations of the surface structure of the finest samples, which cannot be observed at high accelerating voltages.The morphology of the rubber compound is shown in Figure 3.

Surface micrograph in Figure 3 shows that the surface has many tear lines with branching. This pattern indicates the higher the tensile strength of

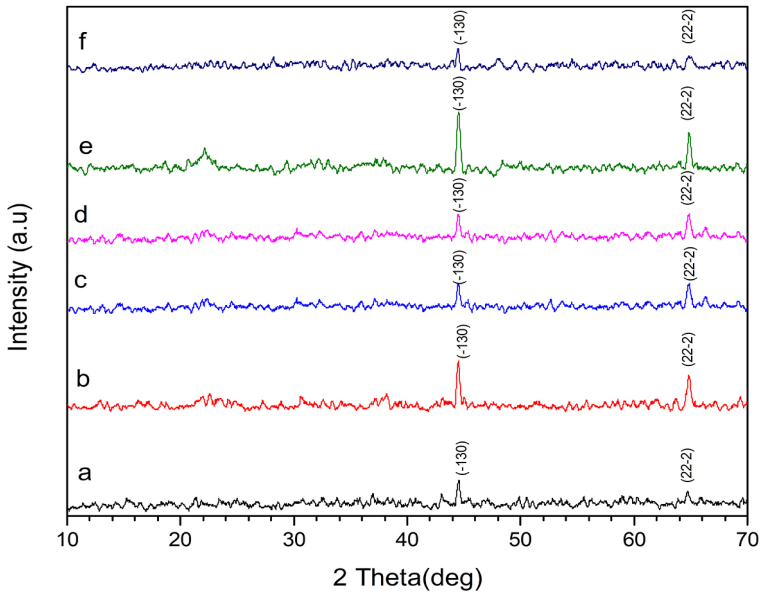

Figure 1. XRD compound analysis with OPBA and bentonite filler: a. OPBA/bentonite (0/100phr), b. OPBA/bentonite (30/70 phr), c. OPBA/bentonite (40/60 phr), d.OPBA/bentonite (50/50 phr), e. OPBA/bentonite (60/40 phr), f. OPBA/bentonite (70/30 phr)

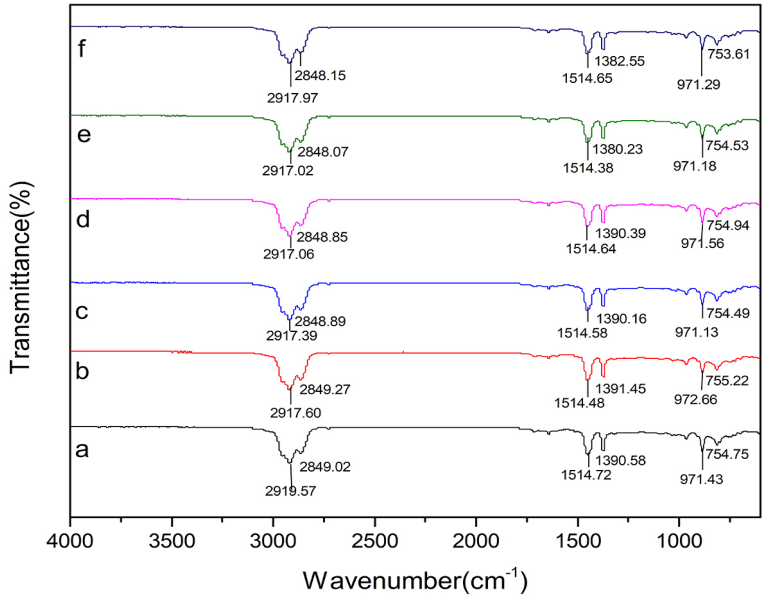

Figure 2. FTIR compound analysis with OPBA and bentonite filler: a. OPBA/bentonite (0/100phr), b. OPBA/bentonite (30/70 phr), c. OPBA/bentonite (40/60 phr), d. OPBA/bentonite (50/50 phr), e. OPBA/bentonite (60/40 phr), f. OPBA/bentonite (70/30 phr)

the compound. All shapes exhibit a rougher surface, indicating that the incorporation of the ultrafine silica filler in the rubber matrix has increased the reinforcement in the rubber vulcanization and has allowed for better stress transfer, consequently increasing tensile strength. Other factors to be considered in reinforcing rubber with fillers, such as: volume fraction, particle size and fillers and filler matrix compatibility (Ginting et al., 2018; Idrus et al., 2011). In addition, Empty cavities arise due to the tendency of silica to form agglomerations because the inner silica has a hydroxyl group, which will help in addition to hydrogen with silica molecules or other polar chemicals.

Mechanical and thermal properties of compound

Tensile strength of rubber compound

The tensile test is a method used to test the strength of a material by applying a force load in the opposite direction. The results from tensile

testing are significant for engineering and product design because they produce material strength data. Tensile test on the rubber compound is shown in Figure 4.

In general, the tensile strength increase with the addition of OPBA/Bentonite filler. It is due to the excellent interaction between the polymer and the hybrid filler system. OPBA/Bentonite is responsible for proper clay dispersion in the rubber matrix. As a result, the load transfer between matrix and filler increases, and the stiffness of the soft rubber matrix. In summary, the improvement in physical properties implies direct evidence for an increase in the contact surface area between the filler and the matrix. As evidenced in SEM micrographs, crack growth resistance increases with the addition of filler (Abhisha et al., 2020; Ginting et al., 2020).

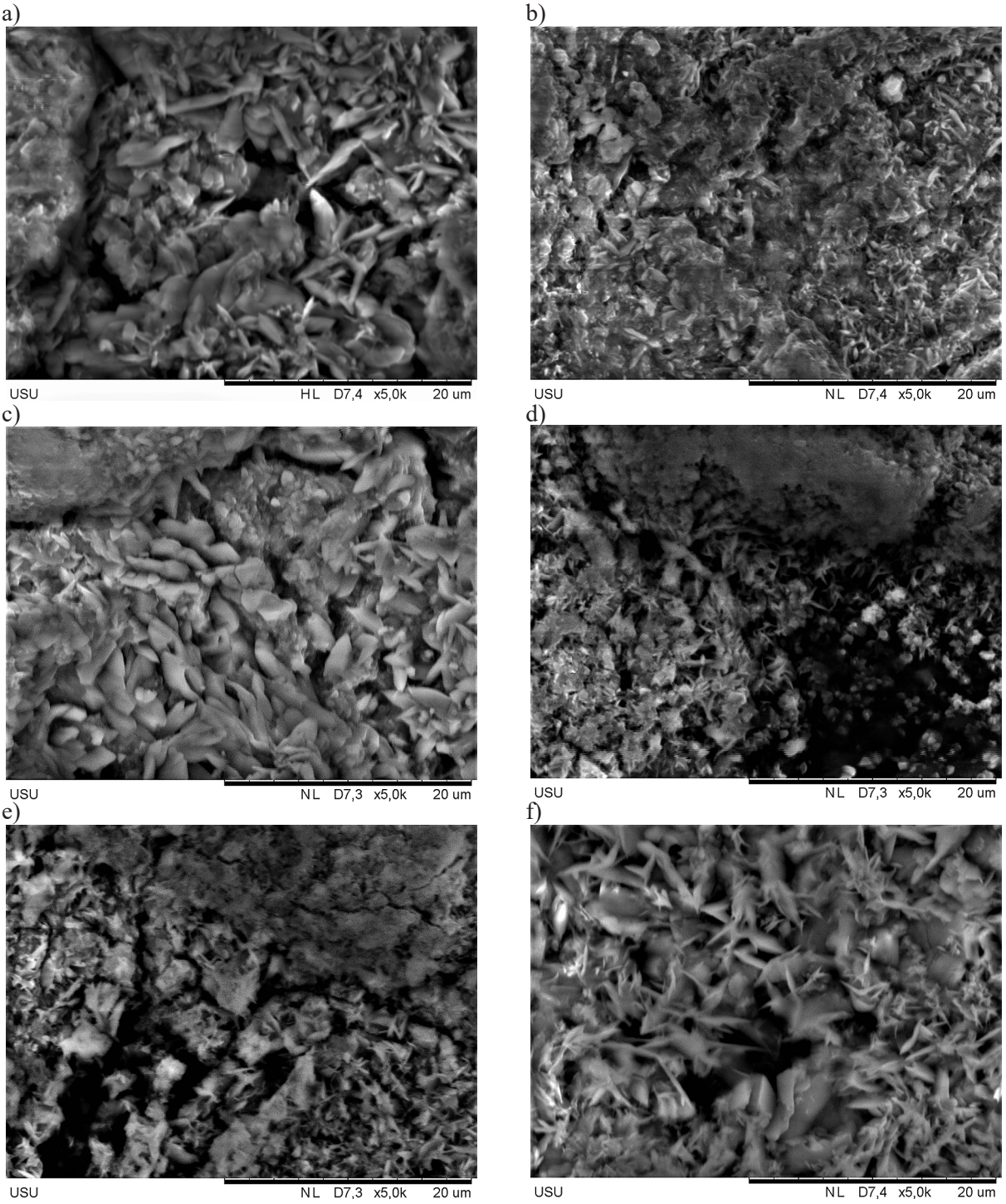

Figure 3. SEM of compound with OPBA/bentonite: a – 0/100 phr, b – 30/70 phr, c – 40/60 phr, d – 50/50 phr, e – 60/40phr, f – 70/30 phr

Elongation at break of rubber compound

The elongation at break is increase in the length of a test piece when stretched to break, expressed by percent of the length of the test piece before stretching. Elongation at break test aims to determine the properties of stress and strains of vulcanized rubber and thermoplastics and including determination of yield point through strength and the increase in the length of the rubber vulcanization when experiencing withdrawals up to a specific extension Furthermore, until it breaks. The Elongation at break of rubber compound with and without filler is shown in Figure 5.

The increase of elongation at break value of rubber compound indicates that a more elastic rubber compound is produced. Elongation at break rubber compound with the most significant value was obtained at 6 wt% filler, which was 110%. Meanwhile, the most negligible elongation at break was obtained at 0 wt% filler, which was 50%. In general, the elongation at break value of the compound increased after the addition of OPBA and Bentonite fillers. Besides, the decreased elongation at break can be caused by increasing cross-linking density between rubber molecules. Elongation at break is one of the physical properties rubber finished goods to determine the elasticity properties of the product that will show up how much product in the form of a ring can be appropriately stretched into place. If the elongation is too high, then the product will be easy to pull so that at its use cannot be tightened with appropriate. The decrease in elongation at break in the filler composition of 8 and 10 wt% is due to the material fillers do not mix homogeneous, so that not all filler can bond with rubber molecules, thereby causing easy vulcanization break when pulled and cause a decrease elasticity (Rahmaniar et al., 2015).

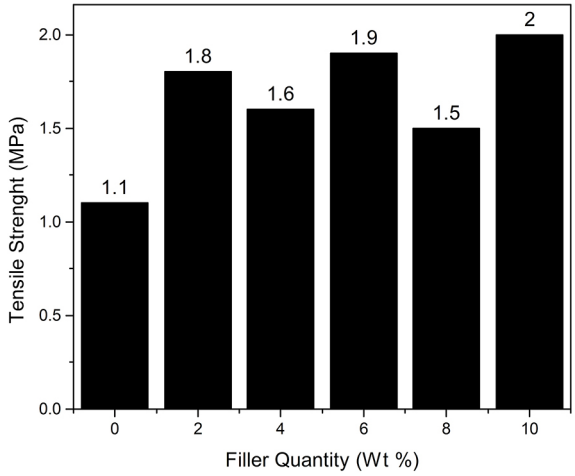

Figure 4. Tensile strenght of compound

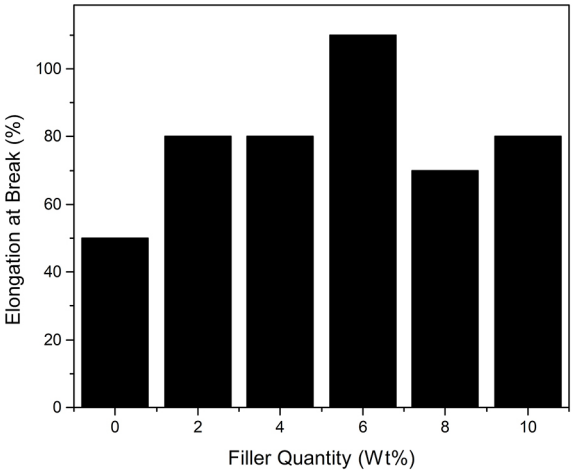

Figure 5. Elongation at break of rubber compound

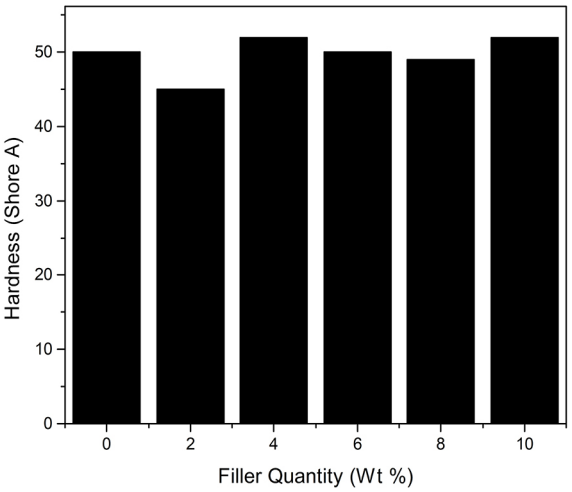

Figure 6. Hardness of rubber compound

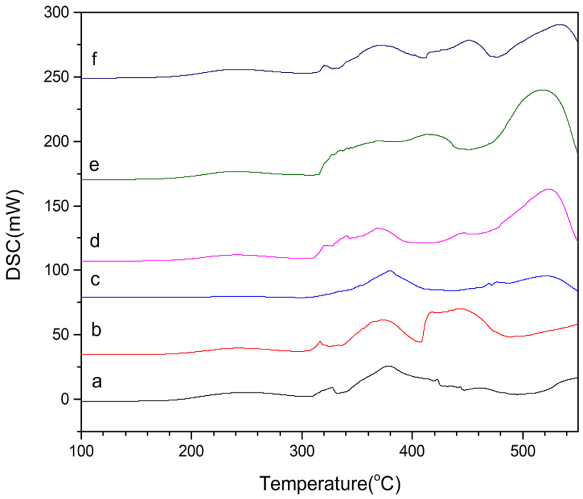

Figure 7. DSC analysis of compound

Table 3. Mechanical properties of rubber compound

Mechanical properties	Unit	Rubber compound					
		S_0	S_1	S_2	S_3	S_4	S_5
Composition filler (OPBA/bentonite)	wt%	0	2	4	6	8	10
Tensile strength	MPa	1.1	1.8	1.6	1.9	1.5	2
Elongation at break	%	50	80	80	110	70	80
Modulus 100%	MPa	-	-	-	1.72	-	-
Hardness	Shore A	50	45	52	50	49	52

Table 4. Differential Scanning Calorimetry data of compound

Peak list	T onset (°C)	T endset (°C)	Peak (°C)
Sample 0			
Peak 1	225.54	259.22	249.55
Peak 2	324.74	329.52	327.10
Peak 3	329.86	343.82	332.51
Peak 4	369.27	386.03	378.51
Peak 5	488.06	505.26	495.16
Sample 1			
Peak 1	221.57	274.57	242.94
Peak 2	321.38	326.95	324.71
Peak 3	354.20	384.38	373.44
Peak 4	438.59	450.31	442.84
Peak 5	563.12	567.54	569.02
Sample 2			
Peak 1	206.73	268.74	241.20
Peak 2	379.74	384.34	379.74
Peak 3	419.62	451.10	434.09
Peak 4	515.51	527.93	520.93
Sample 3			
Peak 1	202.57	270.51	240.43
Peak 2	366.10	378.16	368.69
Peak 3	403.95	419.52	412.57
Peak 4	501.51	539.03	523.72
Sample 4			
Peak 1	213.23	243.34	239.39
Peak 2	411.53	415.49	412.65
Peak 3	448.06	458.21	451.29
Peak 4	512.99	528.98	517.61
Sample 5			
Peak1	209.24	271.88	241.06
Peak 2	352.37	388.91	369.47
Peak 3	405.74	414.35	410.80
Peak 4	444.22	463.51	451.00
Peak 5	469.71	479.36	477.49
Peak 6	527.38	542.11	533.44

Hardness of rubber compound

The hardness value is defined as the resistance that the rubber surface offers against penetration by the indenter and is related to the surface deformation of the rubber surface. The hardness of rubber compound with and without filler is shown in Figure 6.

It can be seen that the trend of change in the hardness of the compound filled with OPBA/Bentonite is similar to the modulus. It can be explained in terms of overall composite stiffness due to crosslink formation and rubber filler interactions (Thongsang et al., 2012). Mechanical properties such as hardness, elongation at break, tensile strength, and Young's modulus of the compounds were analyzed as a function of the weight percentage of the filler material and are presented in Table 3.

Differential scanning calorimetry (DSC) analysis of compound

DSC is a thermoanalytical technique that measures temperature and heat flow and is related to thermal transitions in a material. This thermoanalytical technique measures the difference in the heat required to increase the sample and reference temperatures. Both sample and reference pans have a well-defined heat capacity over the scanned temperature range and are exposed to the same heating history(Kodal et al., 2019). DSC analysis of rubber mixtures can be used to evaluate strengthening effect, and rubber compatibility mixture, as well as rubber-filler interactions. In this study, Shimadzu DSC-60 Plus was used to analyze the thermal properties of rubber compounds. Figure 7 shows Differential Scanning Calorimetry (DSC) Analysis of Compound.

From Table 3, Differential Scanning Calorimetry (DSC) Data of Compound shows that there is an increase in the number of peaks in the sample with 10 wt% filler. Presence of glass transition temperature and melting temperature indicates that the material is semicrystalline and can be processed. In general, polymers are semicrystalline. In other words, some polymer chains are crystalline, and some are amorphous.

CONCLUSIONS

In general, the compounds did not show a change in peak position. Peaks of $2\Theta = 44^{o}$, 64^{o} and dhkl {-130.22-2} were present in all samples. An increase in the amount of filler indicates an increase in intensity. Bonds at 2849–2917 cm-1 were associated with asymmetric methyl stretching vibrations, methyl asymmetric deformation vibrations, and methyl symmetry deformations and the peaks of 1000–650 cm^{-1} showed C = C-H bending. The peak of 1514–1391 cm^{-1} also shows C-H buckling. The compound morphology shows torn lines with branching. Furthermore, in general, the mechanical properties of the compound increased with the addition of OPBA/bentonite filler. This is due to the excellent interaction between the polymer and the hybrid filler system. DSC Compound data showed an increase in the number of peaks in the sample with 10 wt% filler. The presence of a glass transition temperature and a melting temperature indicates that the material is semicrystalline and can be processed.

Acknowledgments

This research was funded by the Directorate of Research, Technology and Community Service, Directorate General of Higher Education, Research and Technology. Ministry of Education, Research Culture, and Technology for Fiscal Year 2022 according to No 059/E5/PG.02.00/PT/2022 and 33/UN5.2.3.1/PPM/KP-DRPM/L/2022

REFERENCES

1. Abdul Salim, Z.A.S., Hassan, A., Ismail, H. 2018. A Review on Hybrid Fillers in Rubber Composites. Polymer - Plastics Technology and Engineering, 57(6), 523–539. https://doi.org/10.1080/03602559.2017.1329432
2. Abhisha, V.S., Augustine, A., Joseph, J., Thomas, S.P., Stephen, R. 2020. Effect of halloysite nanotubes and organically modified bentonite clay hybrid filler system on the properties of natural rubber. Journal of Elastomers and Plastics, 52(5), 432–452. https://doi.org/10.1177/0095244319865573
3. Bao, Z., Tao, J., Flanigan, C. 2015. The Combination of Montmorillonite and Silica in Styrene–Butadiene Rubber/Polybutadiene Rubber Tread Compounds Zuguo. Polymer Composites, 38. https://doi.org/10.1002/pc.23653
4. Bukit, B.F., Frida, E., Humaidi, S., Sinuhaji, P. 2022. Selfcleaning and antibacterial activities of textiles using nanocomposite oil palm boiler ash (OPBA), TiO2 and chitosan as coating. South African Journal of Chemical Engineering, 41(February), 105–110. https://doi.org/10.1016/j.sajce.2022.05.007
5. Bukit, B.F., Frida, E., Humaidi, S., Sinuhaji, P.,

Bukit, N. 2022. Optimization of palm oil boiler ash biomass waste as a source of silica with various preparation methods. Journal of Ecological Engineering, 23(8), 193–199.

6. Bukit, N., Ginting, E.M., Hutagalung, E.A., Sidebang, E., Frida, E., Bukit, B.F. 2019. Preparation and characterization of oil palm ash from boiler to nanoparticle. Reviews on Advanced Materials Science, 58(1), 195–200. https://doi.org/10.1515/rams-2019-0023
7. Farida, E., Bukit, N., Ginting, E.M., Bukit, B.F. 2019. The effect of carbon black composition in natural rubber compound. Case Studies in Thermal Engineering, 16(November), 100566. https://doi.org/10.1016/j.csite.2019.100566
8. Frida, E., Rahmat, F., Bukit, A., Bukit, F. 2022. Analysis structure and morphology of bentonite-Opba. Journal of Applied Research and Technology, 20, 117–125.
9. Ginting, E.M., Bukit, N., Motlan, Saragih, M.T., Frida, E., Bukit, B.F. 2020. Analysis of natural rubber compounds with filler of Oil Palm Empty Bunches Powder and Carbon Black. Journal of Physics: Conference Series, 1428(1). https://doi.org/10.1088/1742-6596/1428/1/012024
10. Ginting, E.M., Motlan, Bukit, N., Saragih, M.T., Sinaga, A.H., Frida, E. 2018. Preparation and Characterization of Oil Palm Empty Bunches Powder. Journal of Physics: Conference Series, 1120(1). https://doi.org/10.1088/1742-6596/1120/1/012004
11. Idrus, S.S., Ismail, H., Palaniandy, S. 2011. Study of the effect of different shapes of ultrafine silica as fillers in natural rubber compounds. Polymer Testing, 30(2), 251–259. https://doi.org/10.1016/j.polymertesting.2010.10.002
12. Ismail, H., Haw, F.S. 2010. Curing characteristics and mechanical properties of hybrid palm ash/silica/natural rubber composites. Journal of Reinforced Plastics and Composites, 29(1), 105–111. https://doi.org/10.1177/0731684408096423
13. Kodal, M., Karakaya, N., Wis, A.A., Ozkoc, G. 2019. Thermal properties (DSC, TMA, TGA, DTA) of rubber nanocomposites containing carbon nanofillers. In Carbon-Based Nanofillers and Their Rubber Nanocomposites: Fundamentals and Applications. Elsevier Inc. https://doi.org/10.1016/B978-0-12-817342-8.00011-1
14. Panitchakarn, P., Wikranvanich, J., Phisalaphong, M. 2019. Synthesis and characterization of natural rubber/coal fly ash composites via latex aqueous microdispersion. Journal of Material Cycles and Waste Management, 21(1), 134–144. https://doi.org/10.1007/s10163-018-0774-x
15. Rahmaniar, Amin, R., Gatot, P., Basuni, H. 2015. Characterization of the rubber compound with extract of caesalpinia sappan wood, quartz sand and shell using. Jurnal Teknologi Industri Pertanian, 25(3), 227–238.
16. Sadhu, S., Bhowmick, A.K. 2004. Preparation and properties of nanocomposites based on acrylonitrile- butadiene rubber, styrene-butadiene rubber, and polybutadiene rubber. Journal of Polymer Science, Part B: Polymer Physics, 42(9), 1573–1585. https://doi.org/10.1002/polb.20036
17. Senthivel, K., Manikandan, K., Prabu, B. 2015. Studies on the Mechanical Properties of Carbon Black/Halloysite Nanotube Hybrid Fillers in Nitrile Rubber Nanocomposites. Materials Today: Proceedings, 2(4–5), 3627–3637. https://doi.org/10.1016/j.matpr.2015.07.118
18. Stöckelhuber, K.W., Das, A., Jurk, R., Heinrich, G. 2010. Contribution of physico-chemical properties of interfaces on dispersibility, adhesion and flocculation of filler particles in rubber. Polymer, 51(9), 1954–1963. https://doi.org/10.1016/j.polymer.2010.03.013
19. Thongsang, S., Vorakhan, W., Wimolmala, E., Sombatsompop, N. 2012. Dynamic mechanical analysis and tribological properties of NR vulcanizates with fly ash/precipitated silica hybrid filler. Tribology International, 53, 134–141. https://doi.org/10.1016/j.triboint.2012.04.006
20. Wang, Z., Liu, J., Wu, S., Wang, W., Zhang, L. 2010. Novel percolation phenomena and mechanism of strengthening elastomers by nanofillers. Physical Chemistry Chemical Physics, 12(12), 3014–3030. https://doi.org/10.1039/b919789c
21. Xu, T., Jia, Z., Luo, Y., Jia, D., Peng, Z. 2015. Interfacial interaction between the epoxidized natural rubber and silica in natural rubber/silica composites. Applied Surface Science, 328, 306–313. https://doi.org/10.1016/j.apsusc.2014.12.029