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Polypropylene-based Multilayer Plastic Waste Utilization on Bitumen Modification for Hot-Mixed Asphalt Application: Preliminary Study

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Abstract

Plastic waste that has not been appropriately managed has caused concern for various stakeholders and has a negative impact on the environment. A way to minimize this issue is by promoting a circular economy through recycling. Despite this, many plastic wastes cannot be economically recycled; one of them is multilayer plastics due to the difficulty in sorting out the various types of materials contained in the layers. As an alternative, it may be possible to utilize multilayer plastic as a bitumen mixture; this method may avoid the separation process entirely. Bitumen is a component of asphalt, which is crucial to the construction of infrastructure. Plastics can potentially enhance the properties of modified bitumen, and they would assist in enhancing the quality of the asphalt. The aim of this study is to examine the impact of multilayer waste plastics on bitumen fabricated by the hot-mixing process. To enhance the compatibility between hydrophobic plastic and hydrophilic bitumen, lignin was used as a compatibilizer. The properties of bitumen with 3, 4, and 5 wt.% multilayer plastic and mixing times of 15, 30, and 45 minutes were evaluated. Accordingly, surface, chemical, thermal properties, and morphology were evaluated using sessile drop tests, Fourier Transform Infrared, thermogravimetry analysis, and scanning electron microscopy.

Keywords: bitumen; compatibility; coupling agent; lignin; multilayer plastic.

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1. Introduction

The plastic industry in Indonesia is growing along with the increasing population growth. Consequently, the need and production of plastics are increasing (Geyer, Jambeck, & Law, 2017; Jambeck, et al., 2015). However, the development of the recycling industry does not accompany it, so there is an increasing amount of plastic waste in Indonesia (Shuker & Cadman, 2018). Multilayer plastic (MLP) is one of the polymer products made of two or more types of plastics that are layered. This type of plastic addresses the need for differences in environmental conditions between the inner and outer layers (Namazi, 2017). Multilayer plastic is in demand to be food wrappers (Ebnesajjad, Plastic films in food packaging, 2013). However, MLP has drawbacks regarding the recycling process due to its lack of ability to segregate. The difficulty of separating the types of material in the layer makes the recycling process can be done very limited (Kaiser, Schmid, & Schlummer, 2017). One alternative is to use multilayer plastic for asphalt mixture to improve the circular economy of multilayer plastic.

Asphalt consists of aggregate and bitumen parts mixed to form a layer and bind to each other. The aggregate consists of rocks or pebbles that function as reinforce, while bitumen is in charge of coating and binding the aggregates so that they become the topmost layer of asphalt. Bitumen is a mixture of sticky organic material and viscous made from hydrocarbons due to oil distillation (Read & Whiteoak, 2003). Bitumen is sensitive to temperature changes, softens when temperatures are high, and tends to be brittle when temperatures are cold. The stress received by bitumen is in the form of dynamic loading of passing vehicles, changes in environmental temperatures, and humidity dynamics due to weather changes (Soenen, Lu, & Laukkanen, 2016). Due to these different types of loading, there is systematic degradation of bitumen quality. Bitumen is a polar material, making this material able to bind to water so that the

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bitumen layer can be eroded. Adding recycled LDPE linear plastic to the bitumen showed good deformation resistance at high temperatures (Nizamuddin, et al., 2020).

The multilayer plastic used for flexible packaging is mainly made of polypropylene or polyethylene, with similar thermomechanical behavior (Wagner, 2016). Using multilayer plastic as a mixture of bitumen to produce a mixture called polymer-modified bitumen (PMB) may solve both problems. The PMB has the advantages of service time and performance of ordinary asphalt due to the thermomechanical and surface properties of plastic (McNally, 2011; (Bulatović, Rek, & Marković, 2011). However, there is challenging to produce high-quality PMB due to the different surface properties of bitumen and multilayer plastic (Pyshyev, Gunka, Grytsenko, & Bratychak, 2016). Bitumen, which tends to be hydrophilic, will be challenging to mix with hydrophobic multilayer plastic appropriately (Zhu, et al., 2014; Ahmedzade, et al., 2015)). Hence, a compatibilizer is needed to facilitate the mixing process to improve the compatibility of bitumen (Yuanita, et al., 2017; Qian, et al., 2018).

Lignin is a phenolic polymeric compound found in lignocellulose from wood in plants (Welker, et al., 2015). Lignin is a by-product of the papermaking process (Biermann, 1998). Lignin is usually used by burning to produce heat that can be used for subsequent production processes (Kim, 2017). Lignin compounds have a unique nature, namely hydrophobic and hydrophilic at the same time (Sannigrahi, Pu, & Ragauskas, 2010). This uniqueness can be used to mix materials with different polarities, in this case, to increase the compatibility between multilayer plastic and bitumen (Xu, Yan, & Zhang, 2015). In this research, a preliminary study has been done to investigate the effect of lignin as a compatibilizer in several types of polymer-modified bitumen mixtures and mixing processes.

2. Methods

The materials used in this research are bitumen pen 60/70 obtained, the multilayer plastic obtained from instant noodle packaging consisting: of oriented polypropylene (OPP) film containing OPP ink, MB Haimaster (PP with organic compound), and OPP layer (Anisa, 2010). Lignin is obtained from the waste of the papermaking process from a local company. Preliminary characterization of the base materials using Fourier Transform Infrared (FTIR), thermogravimetry analysis (TGA), sessile drop tests, and scanning electron microscopy (SEM) were conducted to determine the compatibility between these materials. Multilayer plastics were cut into small pieces and weighed according to the different compositions of PMB, specifically 3, 4, and 5 wt.%. The multilayer plastic pieces were mixed with 500 grams of bitumen and 0.3 wt.% of lignin by hot melt mixing. The PMB mixture was stirred at a temperature of 180 °C for 15, 30, and 45 minutes with 109 rotations per minute. The PMB mixtures were characterized by FTIR, TGA, and SEM.

3. Results and Discussion

3.1. Base Materials Properties

Figure 1a shows the FTIR spectrum of bitumen, where the peaks displayed on the graph represent the groups or chemical bonds possessed by bitumen. The peak of 2921 cm^{-1} indicates the C-H functional group of stretching alkanes. The 2852 cm^{-1} peak indicates the C-H stretching aldehyde functional group. The 1457 cm^{-1} peak shows C-H bending alkanes. The 1375 cm^{-1} peak indicates C-H bending. The peak 723 cm^{-1} peak indicates aromatic and benzene derivatives. The FTIR observation is in accordance with Weigel and Stephan's report (Weigel & Stephan, 2017).

Figure 1b shows the thermogravimetry behavior of bitumen. The decomposition temperature was obtained by calculating the temperature onset, and the result shows that bitumen has a decomposition temperature of 406 °C. This finding is in accordance with the research conducted by J. Mateos that the decomposition of resins, aromatics, and asphaltenes occurs at a temperature range of 350-450 °C (Jiménez-Mateos, Quintero, & Rial, 1996). Bitumen decomposition temperature cannot be determined with certainty due to the influence of the composition of the bitumen constituent material.

The FTIR spectrum of multilayer plastic is shown in Figure 2a. The peaks of 2948 cm^{-1}, 2918 cm^{-1}, and 2866 cm^{-1} indicate alkanes' C-H stretching function groups. The 2836 cm^{-1} peak indicates the C-H functional group of alkanes. Peaks of 1457 cm^{-1} and 1375 cm^{-1} indicate the C-H bending alkane functional groups. The peak of 1182 cm^{-1} indicates the S=O stretching functional group. The C-H stretching function group of alkanes is characteristic of polypropylene polymeric materials. Thermogravimetric was also carried out on multilayer plastic waste, as shown in Figure 2b. The result is that the polypropylene-based multilayer plastic has a decomposition temperature of 368 °C.

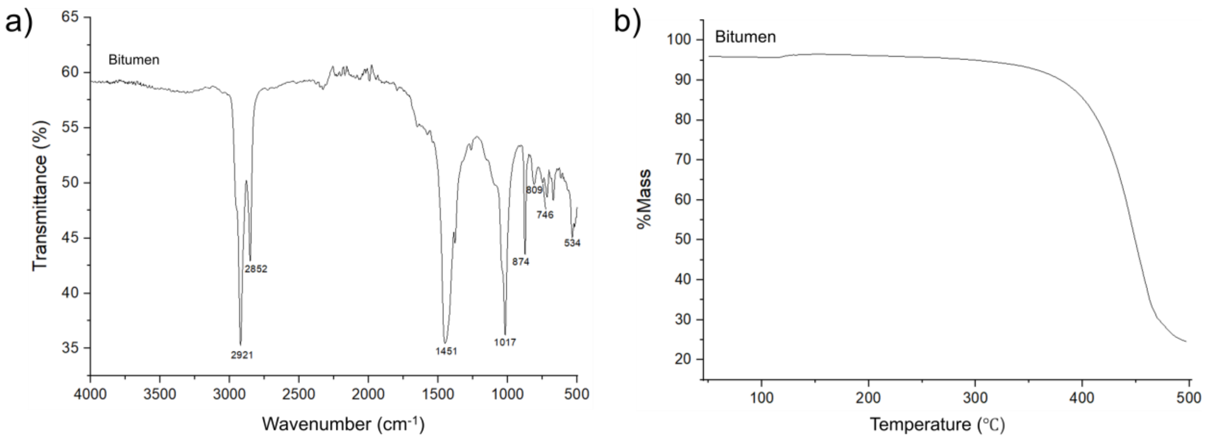

Figure 1. FTIR spectrum (a) and thermogravimetry (b) of bitumen.

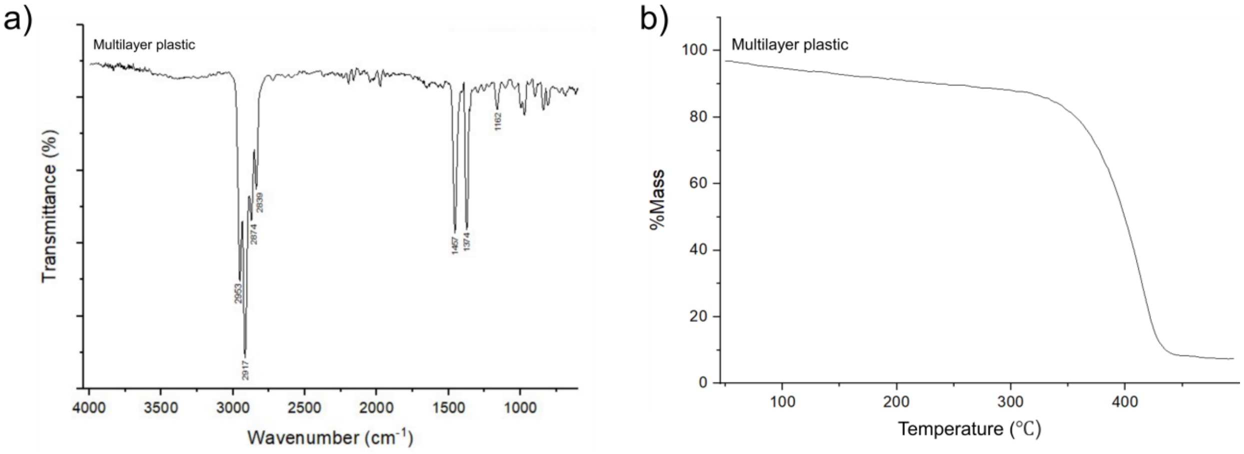

Figure 2. FTIR spectrum (a) and thermogravimetry (b) of multilayer plastic

FTIR spectrum of lignin is shown in Figure 3a. The peak 2918 cm^{-1} and 2851 cm^{-1} indicate the C-H stretching function group of alkanes. The 1606 cm^{-1} peak indicates C=C stretching cyclic alkenes, and the peak of 1028 cm-1 indicates S=O stretching sulfoxide. These confirm the chemical structure of lignin. Thermogravimetry behavior is shown in Figure 3b, where decomposition temperature was observed at 220 °C.

3.2. Materials compatibility

Bitumen and multilayer plastic waste was mixed using the hot melt mixing method to confirm their initial compatibility. The SEM results of mixed bitumen products are shown in Figure 4. although there is a good distribution of SEM results, agglomeration of multilayer plastic was visible. Agglomeration can occur due to the confluence between multilayer plastic particles when stirring is carried out. Plastic particles have the same polarity properties between them, so they tend to stick together.

Figure 5 summarizes the value of the critical surface tension when the surface is entirely wetted, which was obtained from the sessile drop test and calculated by the Zissman method (Ebnesajjad, Surface Tension and Its Measurement, 2014). Bitumen, lignin, and multilayer plastic show a surface tension at 32.3, 29.0, and 25.5 dynes/cm, respectively. The more difference in surface tension value of a material, the more different the polarity properties, making it more difficult to mix. The difference in surface tension corresponds to the value enthalpy. The enthalpy value will be even greater if the difference in surface tension values between the two materials is considerable (Pérez-Lepe, et al., 2006). A significant enthalpy value indicates the non-responsiveness of the two materials to mix. The surface tension value of lignin is between plastic and bitumen, and this indicates the ability of lignin as a compensator in the process of mixing bitumen with recycled plastic (Osa & Good, 1991).

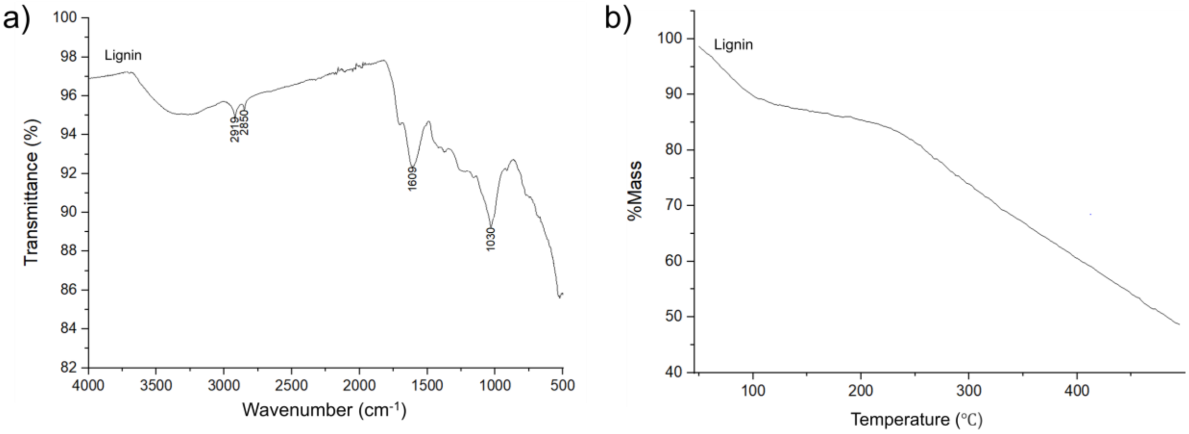

Figure 3. FTIR spectrum (a) and thermogravimetry (b) of lignin.

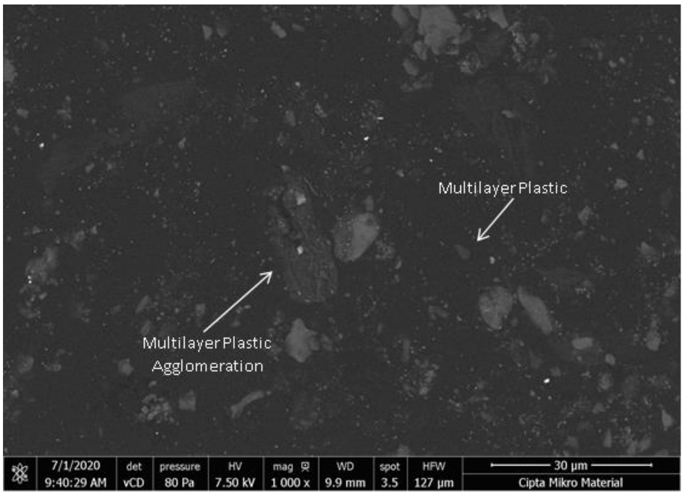

Figure 4. SEM image of polymer modified bitumen without lignin as a modifier (4 wt.% plastic).

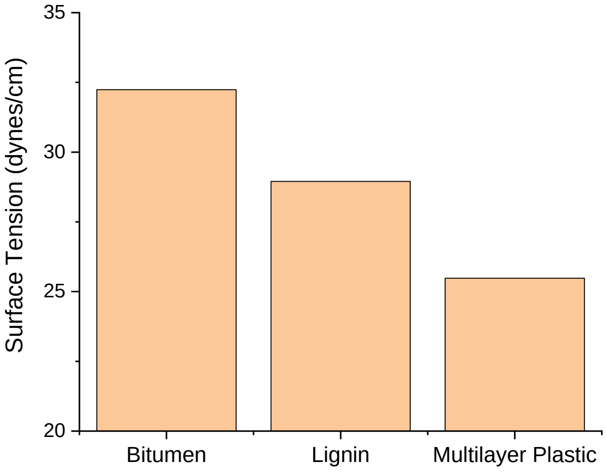

Figure 5. The surface tension of bitumen, lignin, and multilayer plastic.

In addition to the sessile drop test, the compatibility of base materials can also be seen by comparing the peaks in the FTIR spectra between bitumen, plastic, and PMB. Figure 6 shows FTIR spectra of each raw material, where PMB shows the absence of new clusters formed, meaning that bitumen with plastics did not undergo a chemical reaction during the mixing process (Appiah, Berko-Boateng, & Tagbor, 2017). Yuanita reported that the presence of lignin would improve the compatibility between bitumen and polymers, indicated by the disappearance of the -OH functional group in bitumen and lignin. At the same time, a new group of C-O ether hydrogen bonds were reported to be appeared due to the reaction of an -OH functional group of lignin and bitumen (Yuanita, et al., 2017). However, in this study, a new C-O ether bond did not occur. It is likely that the small number of -OH functional groups in bitumen causes lignin to be unable to bind to form the C-O function group of ether, meaning that bitumen with plastic did not react chemically. However, there was a possibility of cohesion between bitumen and polymers (Appiah, Berko-Boateng, & Tagbor, 2017).

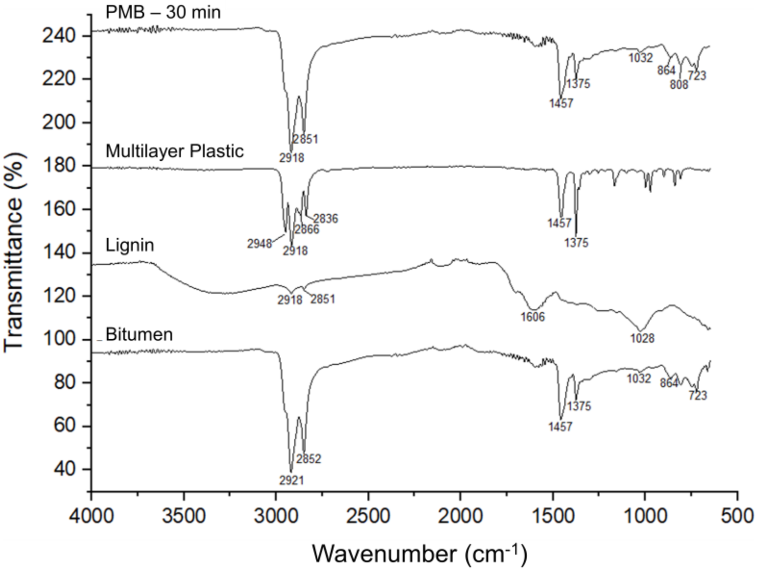

Figure 6. FTIR spectra of bitumen, multilayer plastic, lignin, and PMB

3.3. Influence of Plastic Amount

The effect of the plastic amount on bitumen was studied by comparing PMB with 3, 4, and 5 wt.% plastic. Figure 7 shows that there are no significant changes in the difference in the composition of plastics, which means that there is no chemical interaction formed between plastic and bitumen. These findings are in line with the previous discussion.

The average particle size of plastic was observed by SEM to investigate the influence of plastic composition on its mixing properties. The SEM image can be seen in Figure 8. The average particle size and average closest distance between plastic particles were calculated and summarized in Table 1. It is shown that higher plastic content will lead to a larger average particle size. At the same time, higher plastic content reduces the distance between particles. These findings show that higher plastic content will lead to a worse distribution of plastic. With higher plastic content, the chances of collision between plastic particles become higher, leading to a collision that makes the plastic particles stick to each other and form agglomeration. The agglomeration formed may lead to a decrease in the performance of bitumen as a binder. However, it also may act as a filler which could increase the hardness of bitumen (Tapkın, 2008).

The decomposition temperature of PMB with different plastic content was observed by TGA, and the results were summarized in Figure 9. The decomposition temperature decreases as the plastic composition of the PMB increases. This happens because the plastic material has a lower decomposition temperature than bitumen, leading to a decrease in the decomposition temperature of PMB as the decomposition temperature is highly influenced by the composition of the material (Jiménez-Mateos, Quintero, & Rial, 1996).

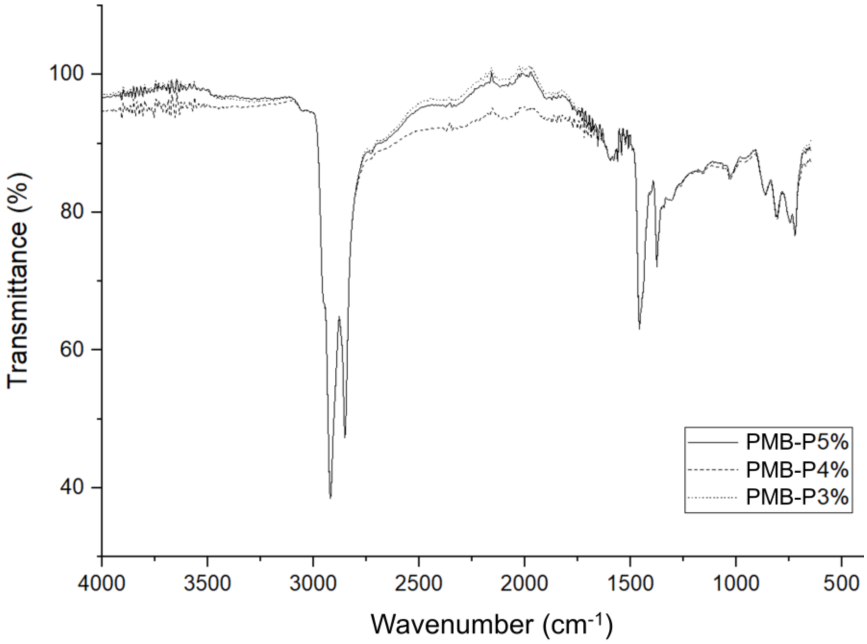

Figure 7. FTIR spectra of PMB with 3, 4, and 5 wt.%

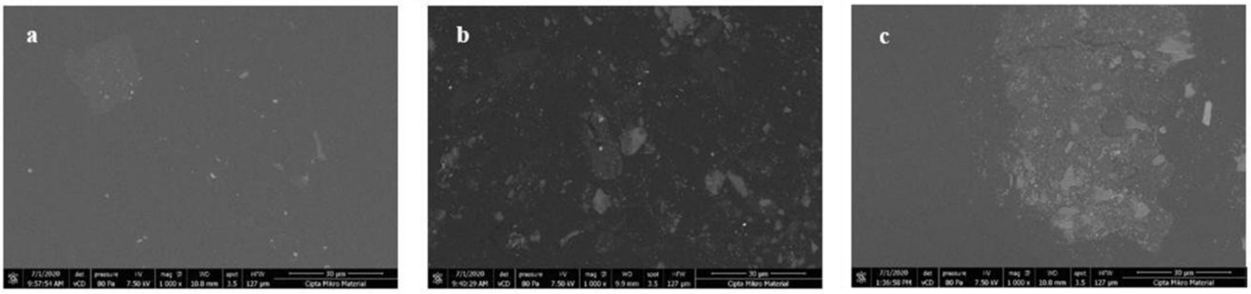

Figure 8. FE-SEM image of PMB with (a) 3 wt.%, (b) 4 wt.%, and (c) 5 wt.% of plastic

Table 1. SEM analysis of PMB with temperature variation

Plastic composition (wt.%)	The average size of particles (µm^2)	The average closest distance between particles (µm)
3	3.2x10^{-4}	1.09
4	5.7x10^{-4}	0.67
5	7.0x10^{-4}	0.34

3.4. Influence of Mixing Time

An investigation of the influence of mixing time has been performed with a PMB containing a multilayer plastic composition of 4 wt.% and lignin composition of 0.3 wt.%, fabricated at 180 °C for 15, 30, and 45 minutes. Figure 10 shows the FTIR spectra of PMB with different mixing times. These spectra demonstrate that no new clusters were formed or were removed. This indicates that there is no chemical reaction from 15 to 45 minutes of mixing. Furthermore, the remaining peaks indicate that neither the plastic nor the bitumen components have been degraded during mixing.

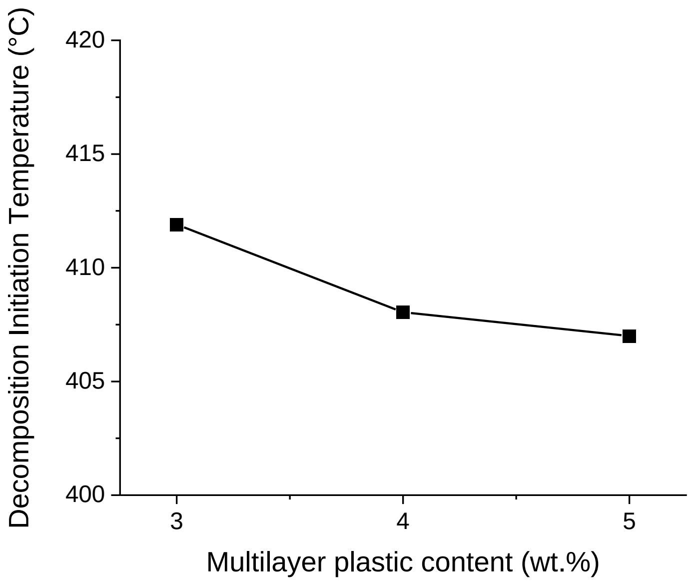

Figure 9. Decomposition initiation temperature of PMB

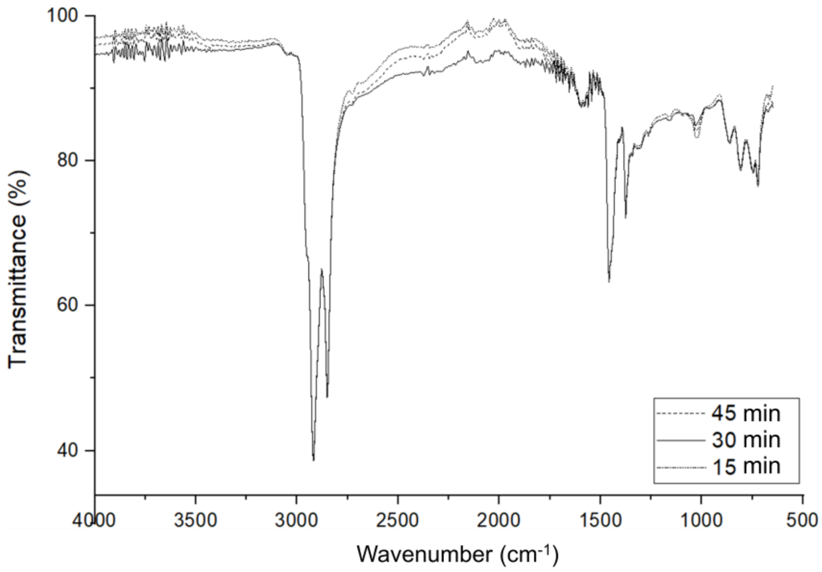

Figure 10. FTIR spectra of PMB with different mixing times.

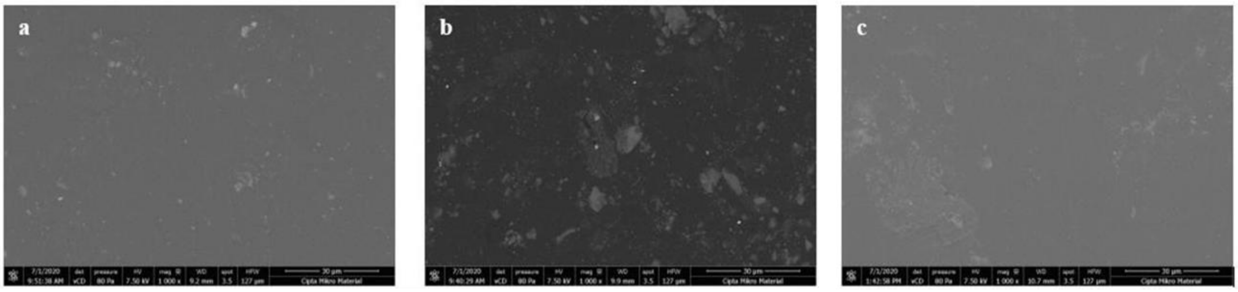

Figure 11. SEM images of PMB with (a) 15 min, (b) 30 min, and (c) 45 min mixing time.

Table 2. Particle size and distribution of PMB with mixing time variation

Mixing time (min)	The average size of particles (μm^2)	The average closest distance between particles (μm)
15	1.7×10^{-4}	4.9×10^{-1}
30	5.7×10^{-4}	6.7×10^{-1}
45	2.0×10^{-4}	6.0×10^{-1}

The morphology of PMB was observed by SEM, as shown in Figure 11. The average particle size and distribution were calculated as summarized in Table 2. The PMB with 15 and 45 minutes shows a smaller average particle size compared to the PMB with 30 minutes, respectively. According to Giavarini, the length of time of stirring will affect the viscosity of PMB, depending on the type of polymer being added (Giavarini, 1994). For thermoplastic polymer types, the viscosity will decrease with increasing stirring time. This can happen due to the friction between the polymer particles, bitumen, and the surface of the machine, which will cause the polymer particles to disperse properly. The larger particle size at 30 min processing time despite the same plastic content means that there is a possibility of an imperfect dispersion of plastic particles so that the size of the plastic particles does not make consistent. The average distance between particles should increase with increasing time as well. However, this is not the case in the 30 minutes PMB sample. Based on research conducted by Gonzalez, there should be an increase in dispersal and distribution properties due to a decrease in viscosity possessed by PMB (Gonzalez, 2010). The more viscosity of a mixture, the greater the ease with which particles will move in it. This causes the presence of plastic particles that move back approaching and tend to agglomerate after dispersion and distribution due to too long a stirring time (Lesueur, 2009). In this study, it is found that 30 minutes of mixing time is enough to obtain optimum dispersion and distribution.

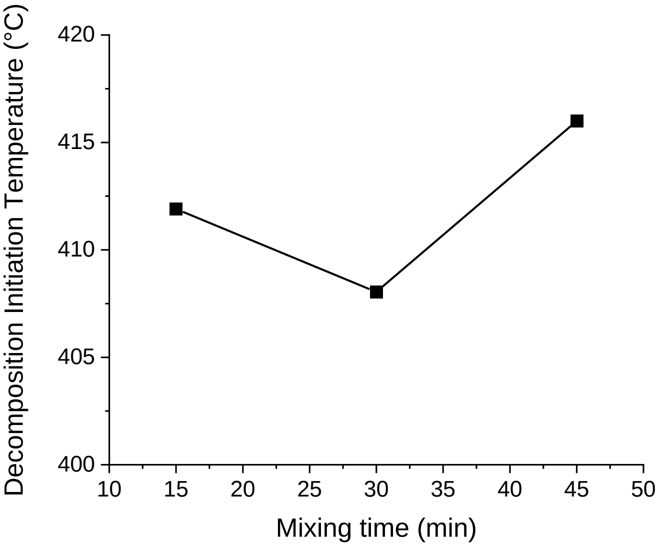

Figure 12. Decomposition initiation temperature of PMB from different mixing times.

Thermogravimetry analyses were performed to evaluate the effect of mixing time on PMB properties. The decomposition temperature was compared and summarized in Figure 12. It is apparent there is a discrepancy between the stirring time and the temperature of the decomposition. In 30 minutes, the temperature marked as low as possible is the temperature recorded at the end of the stirring time. The reason for this might be that the material has been distributed and dispersed more effectively. Re-agglomeration might occur at 45 minutes due to the smaller viscosity, which made it easier for the plastic particles to move so that there was a re-confluence between the dispersed and distributed plastic particles, leading to higher decomposition temperatures.

4. Conclusion

Multilayer plastics have the potential to act as a modifier of PMB mixtures. The multilayer plastic waste is, however, incompatible with bitumen in bitumen mixtures. Lignin may be used as a compatibilizer in these mixtures. In order to create a PMB mixture of the highest quality, process variables must be taken into account, such as the temperature of stirring and the composition of the mixture. By adding more plastic, PMB will be able to agglomerate and have a poorer distribution, especially at 5 wt.% and more. More plastic also lead to decrease PMB thermal properties. The optimum multilayer plastic content is about 4 wt.% of bitumen. A 30-minutes stirring time is enough to obtain the optimum distribution and dispersion. However, advanced improvement is required to improve the PMB quality. It may be possible to achieve this by adding compatibilizers to multilayer plastics or by modifying the plastic in some way so that the bitumen and multilayer plastics become more compatible.

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Contribution: Nugraha: Conceptualization, data analysis, manuscript writing. Naindraputra, Gaol: Data acquisition, data analysis. Ismojo: Design, editing. Chalid: Securing funding, supervision.

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