THE EFFECT OF MULTILAYER PLASTIC WASTE ADDITION TO POLYMER MODIFIED BITUMEN CHARACTERISTICS

Calvin S.A.L. Gaol, Bambang Priyono, M. Chalid, Adam F. Nugraha*
Department of Metallurgical and Materials Engineering, Faculty of Engineering, Universitas Indonesia
calvin.simon01@ui.ac.id
bpriyono@metal.ui.ac.id
chalid@metal.ui.ac.id
adam.febriyanto04@ui.ac.id

ABSTRACT
Bitumen, one of the main components in manufacturing asphalt, is essential for the growing development of infrastructure in Indonesia. Improving the quality of bitumen and the utilization of multilayer plastic waste is the background of this research. Modified bitumen by adding multilayer plastic waste is called Polymer Modified Bitumen (PMB). This study aimed to determine the effect of adding multilayer plastic waste as a bitumen mixture filler. The method used to mix this material is hot melt mixing. The independent variables used were stirring temperatures of 170, 180, and 190 °C, and the composition of plastic waste is 3, 4, and 5 wt%. The characterizations were FTIR, TGA, contact angle test, and SEM. The results showed that the higher the composition level of multilayer plastic waste, the higher the dispersion properties of the mixture and the lower the thermal stability of the mixture.

Keywords: bitumen, multilayer packaging waste, polypropylene, polymer modified bitumen, compatibility

Received *08 June 2022* ***Accepted*** *08 July 2022*

INTRODUCTION

The problem of plastic waste in Indonesia has entered an alarming stage. According to data from the World Bank Group, plastic waste covers 15% of the total solid waste in Indonesia. This problem is getting worse with the total disposal of plastic waste into the oceans of 3.32 million tons and the high percentage of unprocessed plastic waste of 83% in Indonesia [1-2]. A number of studies have been conducted on the utilization and recycling of plastic waste. One of them is the use of plastic waste as a bitumen modifier [3-6].

Bitumen is an asphalt binder made from petroleum and has the following properties: viscoelastic and tends to be hydrophilic or polar. The viscoelastic properties of bitumen cause the mechanical properties of bitumen to be strongly affected by temperature, where bitumen is brittle at low temperatures and easy to creep at high temperatures. Having hydrophilic properties, bitumen will adhere to air, which makes it more prone to deformation. In order to address this shortcoming, polymers are added to bitumen to form Polymer Modified Bitumen (PMB) [7-11].

PMB has been studied mainly through the use of single-layer plastics rather than multilayer plastics since multilayer plastics are more harmful to the environment [12]. The multilayer plastic used in this research is a multilayer plastic made from polypropylene [13]. The PMB mixture showed low compatibility between bitumen and polypropylene-based on previous studies. This is due to the difference in polarity between bitumen and plastic, which causes high interfacial tension between the two components of the mixture. The low compatibility of these two also causes a decrease in the mechanical values of the PMB, such as the ductility value and the penetration value [14]. According to Yuanita et al., the effect of polypropylene addition on the final properties of PMB depends on the composition of the multilayer plastic and the mixing parameters of the bitumen [15].

RESEARCH METHODS

The material used in this research is bitumen pen 60/70 and multilayer plastic in the form of instant noodle wrappers. The instant noodle plastic waste consists of 3 layers, namely: oriented polypropylene (OPP) film plus OPP ink, MB Haimaster (PP with organic compound), and OPP layer [10]. Preliminary characterization of the base materials, such as Fourier Transform Infrared (FTIR) and sessile drop tests, were conducted in order to determine the compatibility between these materials. Multilayer plastics are then cut into small pieces and weighed according to the different compositions of PMB - for example, 3%, 4%, and 5% by weight. The multilayer plastic pieces are mixed with 500 grams of bitumen by hot melt mixing. The PMB mixture was stirred at temperature variations of 170, 180, and 190 °C for 30 minutes with 109 rotations per minute. The PMB mixtures were characterized by FTIR, Thermogravimetric Analysis (TGA), and Field Emission Scanning Electron Microscopy (FE-SEM).

FINDINGS AND DISCUSSIONS

Base Materials Characteristics

Figure 1 shows a graph of the FTIR results from bitumen, where the peaks displayed on the graph show the groups or chemical bonds possessed by bitumen. The peaks of 2921 and 2851 cm^{-1} indicate the C-H bond of the alkane group. The peak of 1595 cm^{-1} indicates the C=C bond of benzene. The peaks at 1450 and 1375 cm^{-1} indicate the $-CH_3$ group. The peak of 1021 cm^{-1} indicates the C-O bond of the carboxylate group. Peaks 874, 809, and 745 cm^{-1} indicate para, meta, and ortho aromatic functional groups [16-17].

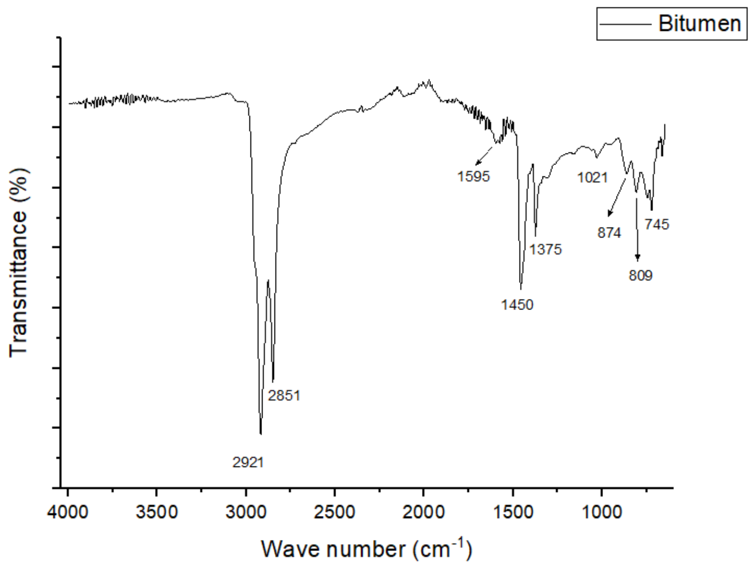

Figure 1. FTIR spectra of bitumen

The result of TGA bitumen in Figure 2 shows the initial decomposition temperature (IDT) of bitumen at 410 °C and maximum decomposition temperature (MDT) at 445 °C.

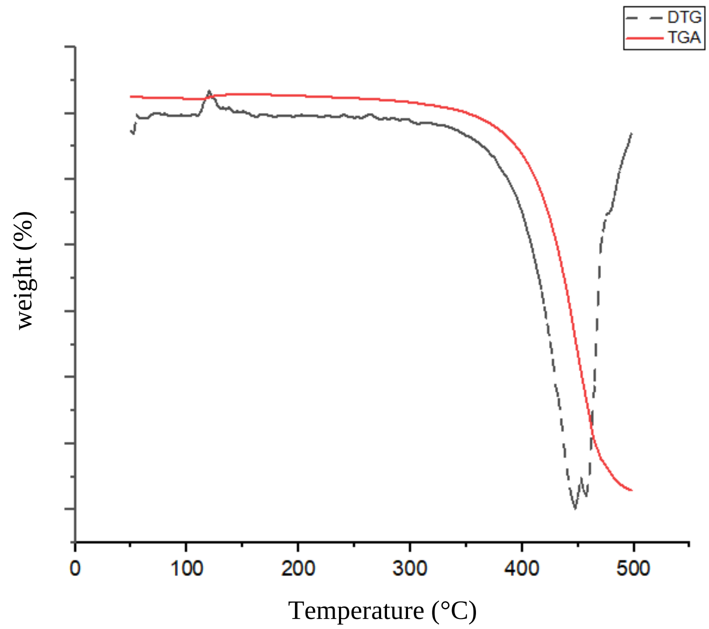

Figure 2. TGA chart of Bitumen

The FTIR spectra in Figure 3 show peaks indicating the types of chemical bonds found in multilayer plastic waste. The peaks of 2953 cm^{-1}, 2917cm^{-1}, and 2874 cm^{-1} indicate the C-H bonds of the alkane chain. The peak of 1457 cm^{-1} and peak of 1374 cm^{-1} indicate the functional group $-CH_3$. The peak of 1162 cm^{-1} indicates the bending of the C-H bond. The peaks of 972 and 842 cm^{-1} indicate the functional groups of aromatic compounds. Aromatic compounds commonly found in plastic packaging waste are derived from anti-radiation additives to protect plastic surfaces from ultraviolet radiation from sunlight [18].

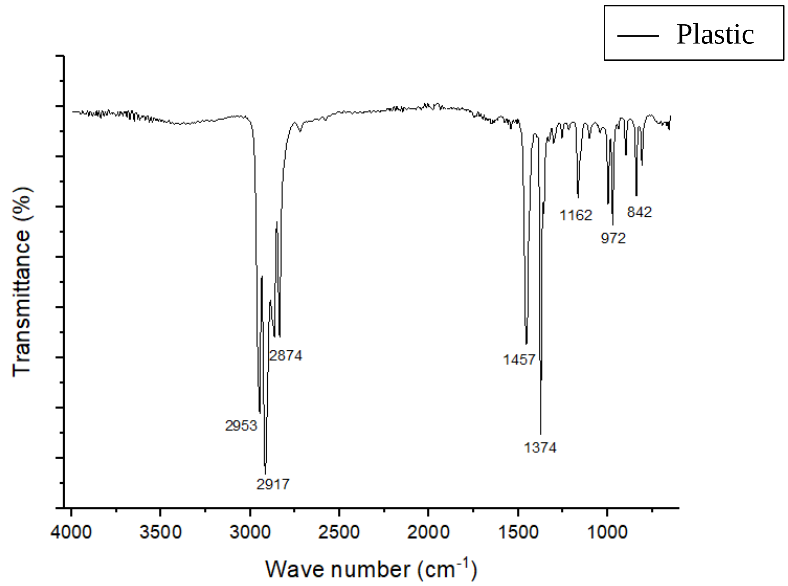

Figure 3. FTIR spectra of Multilayer Plastic

The TGA results for multilayer plastic waste are presented in Figure 4, with IDT for multilayer plastic waste at 365 °C and MDT for multilayer plastic waste at 412.5 °C. These results suggest that PMB has a lower degradation temperature than pure bitumen because multilayer plastic components degrade earlier than bitumen components [19].

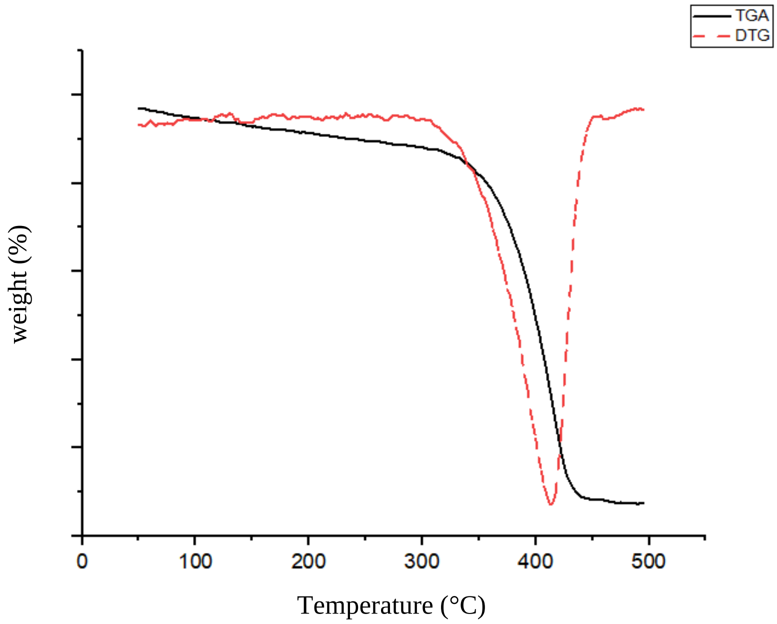

Figure 4. TGA chart of multilayer plastic

Base Materials Compatibility

The sessile drop test was used in order to determine the material compatibility. A sessile drop test was employed to examine the wettability of bitumen as a matrix for wetting PP filler during the mixing of hot melt. The wettability of the compound can be seen from the value of each surface tension possessed by bitumen and water. The surface tension can be calculated using the Zissmann method to analyze the angle results obtained from the sessile drop test [20-21]. The liquid compounds used in this sessile drop test are water and diethylene glycol with a water surface tension of 72 mN/m and diethylene glycol of 48.5 mN/m. The results of the sessile drop test were obtained from the analysis of the contact angle between the liquid droplets and the bitumen and plastic surfaces using ImageJ software. The results of the sessile drop test can be seen in table 1.

Table 1. Base Materials Contact Angle

Base Materials	Liquid	Angle 1	Angle 2	Angle 3	Average
Bitumen	Water	69°	77,1°	75,4°	73,33°
	Diethylene Glycol	49,2°	49,1°	44,2°	47,5°
Plastic	Water	67,7°	64,6°	58,4°	63,56°
	Diethylene Glycol	44,3°	48°	39,7°	44°

Using the contact angle data from table 1, the value of the critical surface tension (surface tension when the surface is entirely wetted) can be determined. The bitumen surface tension value is 31.811 mN/m, and the plastic waste surface tension value is 24.427 mN/m. From the significant difference in surface tension between PP and bitumen, which is 7.384 mN/m, it can be concluded that the compatibility between PP and bitumen is quite poor.

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. The FTIR graph in Figure 5 shows that the PMB FTIR graph shows peaks which are also present in the bitumen and plastic waste FTIR graph. The peaks of 2921 and 2851 cm^{-1} indicate the C-H bond of the alkane group. The peak of 1595 cm^{-1} indicates the C=C bond of benzene. The peaks at 1450 and 1375 cm^{-1} indicate the $-CH_3$ group. The peak of 1021 cm^{-1} indicates the C-O bond of the carboxylate group. Peaks 874, 809, and 745 cm^{-1} indicate para, meta, and ortho aromatic functional groups.

The interaction between bitumen and plastic waste was only shown in the change in %

transmittance at 2921 and 2851 cm-1 (alkane groups addition) and at the peaks of 1450 and 1375 cm^{-1} ($-CH_3$ groups addition). There was no addition or reduction of peaks, which indicated no chemical reaction between bitumen and multilayer plastic waste.

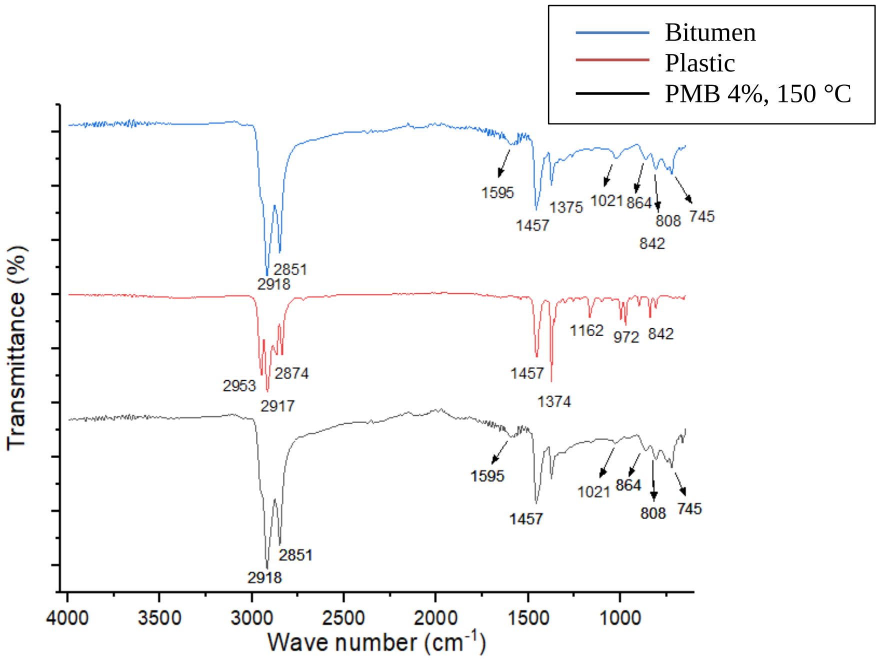

Figure 5. FTIR spectra of Bitumen, Plastic, and PMB

The Effect of Mixing Temperatures and Plastic Composition

The FE-SEM test was performed to determine the effect of mixing temperature on the compatibility of PMB components. The morphological results of the PMB surface are shown in Figures 6 and 7. The dispersion and distribution analysis results are shown in Tables 2 and 3. By temperature variation, the best dispersion (smallest particle size) was at ≤ 170 °C, and the best distribution (highest closest distance) was at 180 °C. The higher the stirring temperature, the lower the viscosity of the bitumen and plastic so that the plastic particles will spread easier in the bitumen mixture. However, at high temperatures, the tendency of plastics to agglomerate is also higher. Therefore, it is necessary to experiment to determine the optimal stirring temperature. By plastic composition variation, the best dispersion (smallest particle size) is 5 wt%, and the best distribution (highest closest distance) is 3 wt%. The higher the polymer composition in the PMB mixture, the more difficult it is for particles to be evenly dispersed [22]. This is due to the poor compatibility between bitumen and multilayer plastics, which causes polymer particles to tend to agglomerate. Considering that too small a particle size can cause a decrease in mechanical strength, it can be concluded that the optimum mixing temperature is 180 °C, and the optimum plastic composition is 3 wt% [13].

Table 2. Table of SEM analysis of PMB with temperature variation

Temperature (°C)	Average size of particles (μm^2)	Average closest distance between particles (μm)
170	6.88×10^{-2}	5.12×10^{-1}
180	1.16×10^{-1}	9.52×10^{-1}
190	1.41×10^{-1}	7.13×10^{-1}

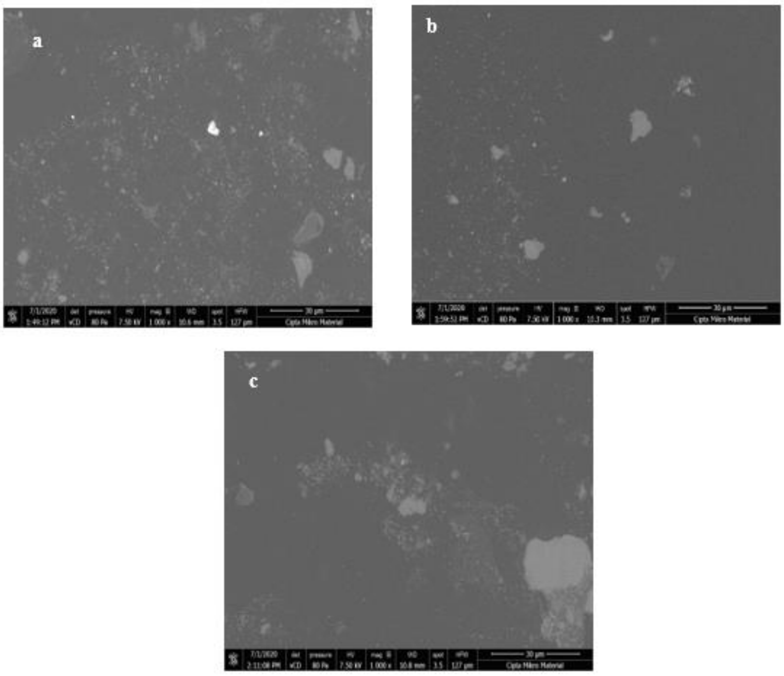

Figure 6. FE-SEM image of PMB at (a) 170 °C, (b) 180 °C, and (c) 190 °C

Table 3. Table of SEM analysis of PMB with temperature variation

Composition (wt%)	Average size of particles (μm^2)	Average closest distance between particles (μm)
3	4.47×10^{-2}	7.19×10^{-1}
4	1.16×10^{-1}	9.52×10^{-1}
5	1.73×10^{-2}	1.61

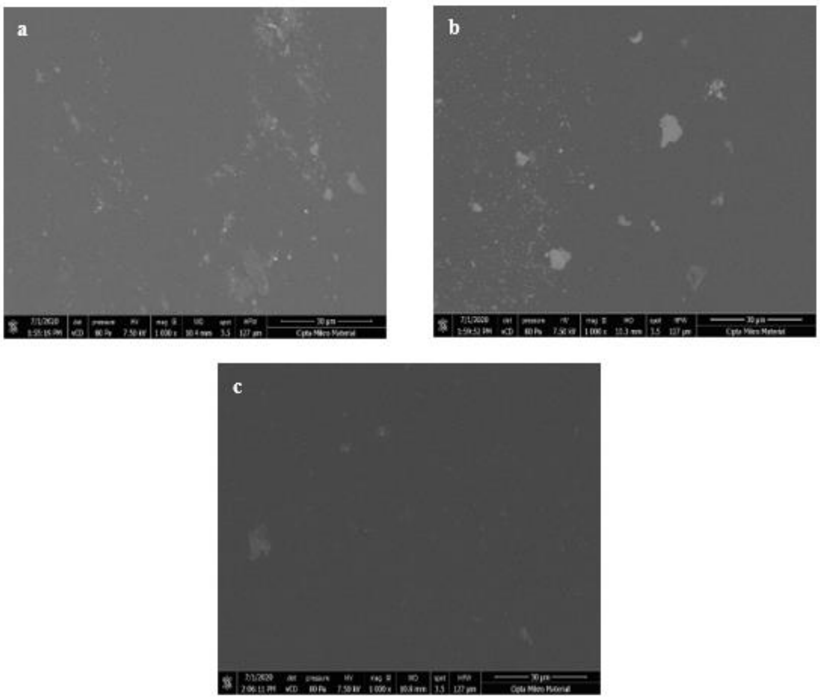

Figure 7. FE-SEM image of PMB at (a) 3 wt%, (b) 4 wt%, and (c) 5 wt%

CONCLUSION

Multilayer plastic has the potential to be a modifier in PMB mixtures. In creating a PMB mixture of the highest quality, various process variables must be considered, such as the stirring temperature and the composition of the mixture. The test results show that the optimum mixing temperature for PMB with multilayer plastic is 180°C and the ideal multilayer plastic composition is 3 wt% of bitumen. A further improvement in this quality can be achieved by increasing the compatibility between bitumen and multilayer plastic through either adding compatibilizers or modifying the plastic in some way.

REFERENCES

[1] I. Shuker and C. Cadman, Indonesia - Marine Debris Hotspot Rapid Assessment: Synthesis Report, World Bank (2018).

[2] J. R. Jambeck, R. Geyer, C. Wilcox, T. R. Siegler, M. Perryman, A. Andrady, R. Narayan dan K. L. Law. (2015). "Plastic Waste Inputs from Land into the Ocean," Science Mag, 347(6223), p. 768-771

[3] F.Knappich, F. Hartl, M. Schlummer, & A. Mäurer. (2017). Complete Recycling of Composite Material Comprising Polybutylene Terephthalate and Copper. Recycling, 2(2), 9. https://doi.org/10.3390/recycling2020009

[4] S. Hinislioglu. (2004). Use of waste high density polyethylene as bitumen modifier in asphalt concrete mix. Materials Letters, 58(3-4), 267-271. https://doi.org/10.1016/s0167-577x(03)00458-0

[5] L. Brasileiro, F. Moreno-Navarro, R. Tauste-Martínez, J. Matos, M.C. Rubio-Gámez. (2019). Reclaimed Polymers as Asphalt Binder Modifiers for More Sustainable Roads: A Review. Sustainability 2019, 11, 646

[6] Selpiana, P. Susmanto, S. Miskah, L. Dharmawan, dan M. F. R. Akbar. (2019). Pemanfaatan Limbah Plastik Berlapis Alumunium (Multilayer) dengan Metode Solvasi. Palembang : Fakultas Teknik Kimia Universitas Sriwijaya.

[7] V. Bulatović, V. Rek and K. Marković. (2015) " Polymer modified bitumen," Materials Research Innovations, vol. 16, no. 1, pp. 1-6.

[8] T. McNally, Polymer Modified Bitumen, Oxford: Woodhead Publ., (2011).

[9] S. Tapkın. (2008). The effect of polypropylene fibers on asphalt performance. Building And Environment, 43(6), 1065-1071. https://doi.org/10.1016/j.buildenv.2007.02.011

[10] C. Giavarini. (1994). Chapter 16 Polymer-Modified Bitumen. Developments In Petroleum Science, 381-400. https://doi.org/10.1016/s0376-7361(09)70263-8

[11] P. Ahmedzade, K. Demirelli, T. Günay, F. Biryan, O. Alqudah. (2015). Effects of Waste Polypropylene Additive on the Properties of Bituminous Binder. Procedia Manuf. 2015, 2, 165–170

[12] K. Kaiser, M. Schmid and M. Schlummer. (2017). "Recycling of Polymer-Based Multilayer Packaging: A Review," Recycling, vol. 3, no. 1, (2017) p. 1.

[13] A. Anisa. (2010). "Evaluasi dan Analisis Waste pada Proses Produksi Kemasan Menggunakan Metode FMEA (Skripsi)," Universitas Indonesia.

[14] J. Zhu, B. Birgisson and N. Kringos. (2015). "Polymer modification of bitumen: Advances and challenges," European Polymer Journal, vol. 54, pp. 18-38.

[15] E. Yuanita, B.E. Hendrastyawan, F.D. Firdaus, & M. Chalid. (2017). Improvement of polypropylene (PP)-modified bitumen through lignin addition. IOP Conference Series: Materials Science and Engineering, 223, 012028.

[16] R. Ren, K. Han, P. Zao, J. Shi, L. Zao. (2018). Identification of Asphalt Fingerprint and Principal Component-Liniar Discriminant Analysis. Construction and Building Materials.

[17] S. Weigel, & D. Stephan. (2017). The prediction of bitumen properties based on FTIR and multivariate analysis methods. Fuel, 208, 655-661. https://doi.org/10.1016/j.fuel.2017.07.048

[18] L. Barbes, C. Radulescu, C. Stihi. (2014). ATR-FTIR Spectrometry Characterisation of Polymeric Materials. Romanian Reports in Physics, Vol. 66, No. 3, p. 765–777

[19] J. Jiménez-Mateos, L. Quintero, & C. Rial. (1996). Characterization of petroleum bitumens and their fractions by thermogravimetric analysis and differential scanning calorimetry. Fuel, 75(15), 1691-1700. https://doi.org/10.1016/s0016-2361(96)00169-x

[20] G. Lamour, A. Hamraoui, A. Buvailo, Y. Xing, S. Keuleyan, & V. Prakash. et al. (2010). Contact Angle Measurements Using a Simplified Experimental Setup. Journal Of Chemical Education, 87(12), 1403-1407. https://doi.org/10.1021/ed100468u

[21] S. Ebnesajjad. (2014). Surface Tension and Its Measurement. Surface Treatment Of Materials For Adhesive Bonding, 7-24. https://doi.org/10.1016/b978-0-323-26435-8.00002-2

[22] C. Gogos, & Tadmor,Z. (2006). Principles Of Polymer Processing, Second Edition. John Wiley & Sons, Inc., Publication.