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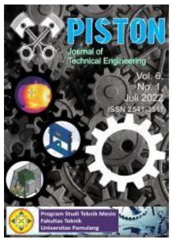

Effect of Cold Plasma Treatment on Physical Properties of Multilayer Plastics for Polymer Asphalt Applications

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Abstract: Multilayer plastic waste continues to increase due to the ever-growing consumption and needs of the global citizen and is one of the most challenging types of waste to recycling because of its nature. The accumulation and indiscriminate disposal of waste can pose a potential risk of environmental problems. A solution that can be implemented is to mix bitumen and waste polymer as asphalt manufacturing. Despite its advancement in research, many potential parameters are still to be discovered to achieve optimal results. Through cold plasma treatment, a surface treatment may occur at the multilayer polypropylene that causes the change from hydrophobic to hydrophilic properties. Treating the polymer with cold plasma will provide good hydrophilic properties without changing the overall chemical and thermal properties of the sample. This will result in an alternative aggregate for the bitumen for asphalt manufacturing. Bitumen coupled with the addition of plastic still provides an optimal hardness and ductility, meanwhile having a more economically viable manufacturing process than other processes.

Keywords: Multilayer Plastic, Bitumen, Cold Plasma, Modified Bitumen, Hydrophilic Surface.

Abstrak: *Sampah plastik berlapis terus meningkat karena konsumsi dan kebutuhan warga dunia yang terus meningkat dan merupakan salah satu jenis sampah yang paling menantang untuk didaur ulang karena sifatnya. Penumpukan dan pembuangan limbah yang sembarangan dapat menimbulkan potensi risiko permasalahan lingkungan, solusi yang dapat diterapkan adalah dengan mencampurkan bitumen dan limbah polimer sebagai pembuatan aspal. Meskipun kemajuannya dalam penelitian, banyak parameter potensial yang masih harus ditemukan untuk mencapai hasil yang optimal. Melalui perlakuan plasma dingin, perlakuan permukaan dapat terjadi pada polipropilena multilayer yang menyebabkan perubahan sifat hidrofobik menjadi hidrofilik. Dengan perlakuan polimer dengan plasma dingin akan memberikan sifat hidrofilik yang baik tanpa mengubah sifat kimia dan termal keseluruhan sampel. Ini akan menghasilkan agregat alternatif untuk aspal untuk pembuatan aspal. Bitumen ditambah dengan penambahan plastik masih memberikan kekerasan dan daktilitas yang optimal, sementara memiliki proses manufaktur yang lebih ekonomis daripada proses lainnya.*

Kata kunci: *Plastik Berlapis, Bitumen, Plasma Dingin, Bitumen Modifikasi, Permukaan Hidrofilik.*

INTRODUCTION

Plastic is an inseparable and essential part of human life. Plastics have been widely used in all life, from plastic products to daily necessities for tableware to the wide application of the latest technological products, such as the connection between materials and semiconductor materials [1]. The wide use of plastic is inseparable from the two advantages of plastic, namely its nature and its simple and inexpensive production process that can be changed according to needs [2].

Among the many plastics produced, the packaging industry is the industrial sector that produces the most plastic. From the study by Gail et al. in 2017, it was found that the packaging industry produced 146 million tonnes of plastic per year in 2015 [1]. However, due to the short life span of a single package, of the 146 million tons of plastic produced in the packaging industry, 141 million tons ended up being a plastic waste.

Recycling is an alternative that can be used to overcome environmental pollution because recycling can reduce the amount of plastic waste in the world [5]. Simply put, recycling is one of the most effective ways to protect and preserve the nature in which we live. By reusing old materials, we save the energy needed to make new ones [5-6].

One of the things that have an impact on the reuse of old materials is the circular economy sector [4]. The circular economy concept is guided by the principle of reducing waste and maximizing existing resources. This circular economy approach is different from the traditional linear economy, which uses a take - use - dispose model (take - make - dispose). In a circular economy system, the use of resources, waste, emissions and wasted energy is minimized by closing the production-consumption cycle by extending product life, design innovation, maintenance, reuse, remanufacturing, recycling to the original product (recycling), and recycling into other products (upcycling). In the context of the sustainability of plastic products, the circular economy concept can be applied in several ways: recycling plastics, upcycling plastics as a mixture of asphalt, converting plastics of low economic value into fuel or energy, and so on [4].

The most common type of plastic packaging in Indonesia is polypropylene (PP) [3]. The general characteristics of polypropylene are good impact resistance, high chemical resistance, and high rigidity and tensile strength. One type of PP-based plastic packaging is sheet plastic (film), widely used as food packaging. Most of these plastic sheets are multilayer, and single-use types [5]. The system boundary for multilayer food packaging can be seen in Fig.1. One of the goals of the multilayer is to make plastic packaging more resistant to tearing, with better heat resistance, airtightness, and water resistance [6, 7]. Unfortunately, in Indonesia, the recycling value of multilayer plastic types (such as instant noodle wrappers) is usually much lower than that of PP plastics. This is because multilayer plastics have different characteristics and melting points between materials and are less attractive to scavengers [7]. As a result, the recycling process is more time-consuming and more expensive than usual.

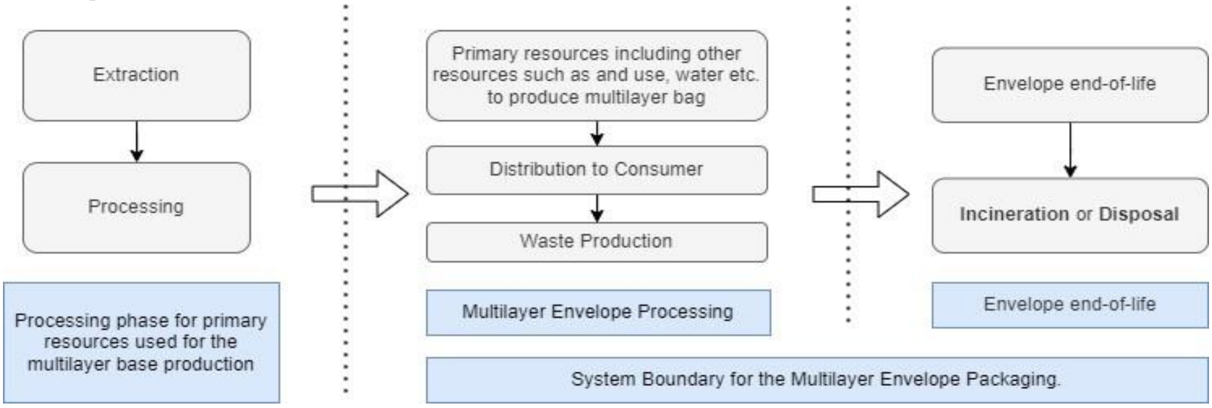

Figure 1. System Boundary for Multilayer Food Packaging (5)

Several researches on polymer asphalt have emerged and have even been applied. However, there are still problems that arise from such a breakthrough. One of them is a polymer that is still difficult to mix with the asphalt or bitumen composition itself. The chemical dissimilarity of bitumen and polymer usually gives two different phases in the mixing mechanism, namely the polymer phase and the bitumen phase [13]. The polymer phase is formed by the development between the chains at the lowest intensity and bitumen compatibility, while the bitumen phase is formed by the concentrated weight fraction (mainly tin content) [13]. The nature of the network or chain structure of the polymer and its effect on polymer modification is a function of the basic properties of the asphalt, the properties, and content of the polymer, and the suitability of the polymer asphalt softness (polymer asphalt stiffness) and increased permeability index (PI) [14,16].

Another problem found in the nature of the plastic is "water-resistant," or can be known as hydrophobic [17]. The plastic that is a mixture of asphalt blocks the absorption of water so that water will stagnate and erode the asphalt surface over time. This makes asphalt less durable and requires frequent maintenance [18]. From these problems, what researchers should do in future research is to change the nature of the plastic which is used as a mixture of asphalt or bitumen. Therefore, the plastic used as an asphalt mixture should be hydrophilic in order to absorb water from the road and not stagnate, which will interfere with traffic. With hydrophilic properties, water will seep and penetrate the asphalt base. It is better if a tiny channel between the asphalt and concrete of the road foundation directs water to the edge and is made into a sewer. In order to improve other applications of the material and without compromising the relationship of most of the polymer asphalt chains, modifications to the polymer chain structure are required, which can change the properties of the plastic surface used. Plastic surface modification is significant in bituminous mixtures. It is hoped that changing the plastic's properties can increase the absorption capacity better than the previous polymer asphalt [18-21].

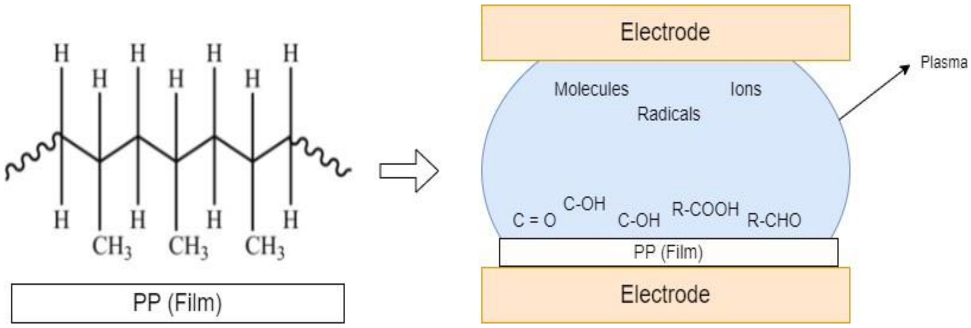

Figure 2. Irradiation of Plasma for Polypropylene Film Surface Treatment

METHODS

One of the breakthroughs that can be done is to use plasma to engineer the surface of multilayer plastics used as bitumen mixtures. Plasma treatment is the most popular polymer surface modification process [22]. Plasma modification is clear, reacts quickly, produces low pollution and good coating, and can be engineered from the polymer chain structure. During the plasma excitation process, atoms, ions, radicals, and UV rays are produced in the plasma [22]. Plasma can improve properties such as permeability, conductivity, printability, adhesiveness, or biocompatibility of the polymer surface [23]. One method of plasma that is quite environmentally friendly is cold plasma or cold plasma [24].

Table 1. Material Composition of Multilayer Plastic

Materials	
Main Materials	OPP *Film*
	PP Cosmoplene
	MB Haimaster White
Ink Materials	OPP *Red*
	OPP *Green*
	OPP *Medium* GOPP
	OPP *White* GOPP
	OPP *Yellow* GOPP
	OPP *Grey* GOPP
	OPP *Blue*
	PPL *Black*
Solvent Materials	*Ethyl Acetate*
	Methyl Ethyl Ketone
	Toluene
Other	*Blank Paper Core*

Cold plasma firing is a vital modification technique in modern industry. This is because cold plasma uses the reactive gas principle, which can change the surface characteristics of the material or product without changing the overall characteristics of the material or product [24]. This study aimed to develop a cold plasma system operating at atmospheric pressure and to study the effect of cold plasma treatment on the surface properties of polypropylene (PE) polymeric materials under varying time conditions. The use of organic polymer plasma is also a popular plasma modification method [24], one of which is cold atmospheric plasma, which utilizes air or the earth's atmosphere without a vacuum [25] and therefore, in this study, an experiment will be conducted using this method, especially for changing the hydrophobic to hydrophilic properties of plastics.

Materials such as PEN 60/70 asphalt, plastic laminate, and lignin were used in this study. The multilayer plastic used in this study consisted of 150 instant noodle plastic waste from various sources. The multilayer plastic has some composition for each layer, as shown in Table 1. This waste is adequately collected to avoid contact with environmental pollutants such as soil.

Preparation of Modified Bitumen

1. Shredding of used multilayer plastic

The multilayer plastic is still too heavy and bulky to execute. It is difficult to get a soft and uniform dough with this large volume. For small and uniform particles, counting is required. To reduce the size of the plastic, cut it with scissors and then cut it with a grinder.

2. Multilayer plastic modification

The plastic that had been chopped into small pieces was washed with clean water and then dried for about 1 hour. Then the plastic is put into a cold plasma reactor with a power of 15 W, a voltage of 10 V, and a current of 15 mA to modify the properties of the plastic with the variables mentioned in Figure 3. After getting the results, some of the modified plastics are used as samples to be tested for characterization, and the rest will be mixed with bitumen by hot melt mixing.

3. Mixing of bitumen and multilayer plastic

The next preparation is mixing bitumen with plastic that has been treated with plasma and has been chopped. The total weight of the sample that is inserted into the chamber is 500gr. Then given, variables in the form of the amount of plastic content, duration of treatment, and samples that did not use a multilayer plastic mixture. Then the chamber is heated in a mixing machine to a temperature of 110^{o}C, which is then mixed at a speed of 109 rpm for 30 minutes. Then the chamber is removed from the mixing machine, and then the sample is poured from the chamber into a can which is used as a container for storing samples and making it easier for testing mobility. To facilitate the analysis, each sample is given an abbreviation in the form of a sample code and summarized in a table. The table can be seen in Table 2.

Table 2. Sample Code from Each Variable

Number	Sample Code	Description
1	BP_0	Pure Bitumen
2	BP_1T_0	Bitumen + 1% untreated plastic
3	$BP_{2,5}T_0$	Bitumen + 2.5% untreated plastic
4	BP_5T_0	Bitumen + 5% untreated plastic
5	BP_1T_{30}	Bitumen + 1% of treated plastic with 30 second plasma treatment
6	$BP_{2,5}T_{30}$	Bitumen + 2.5% of treated plastic with 30 second plasma treatment
7	BP_5T_{30}	Bitumen + 5% of treated plastic with 30 second plasma treatment
8	BP_1T_{60}	Bitumen + 1% of treated plastic with 60 second plasma treatment
9	$BP_{2,5}T_{60}$	Bitumen + 2.5% of treated plastic with 60 second plasma treatment
10	BP_5T_{60}	Bitumen + 5% of treated plastic with 60 second plasma treatment

Characterization

1. Contact Angle Test

The contact angle test (sessile drop test) aims to determine the wettability of a material. This test is carried out by measuring the contact angle of a liquid whose standard surface tension is known on the surface of the raw material, which is then measured by the contact angle of the liquid droplet.

2. Chemical Composition Test

FTIR (Fourier Transform Infrared Spectroscopy) is based on the vibration of a molecule. The principle of FTIR is to look at the interaction of energy that comes from vibrations with a material.

3. Mechanical Testing

A. Ductility Test

This test is carried out to determine the maximum strain value that the material will experience before the material breaks. This test procedure is adjusted to the standard ASTM D-113-99 or SNI-06-2432-1991. The procedure is carried out as follows:

- Heating the sample with some of the temperatures in 120°C-150°C until it melts and then stirs until smooth
- Putting the sample into the mold until it dries
- Soak the sample in water at 25°C for 1 hour
- Withdraw the sample at a speed of 50 mm/min until the sample breaks

B. Penetration Test

This test is performed to determine the strength of the asphalt, assessed by a penetration detector. Bitumen can be classified as high strength bitumen if the penetration path is low. During the invasion, the bitumen becomes less strong. The sample used for this test is a standard temperature sample that does not require special handling. This test is performed by inserting a needle into the sample at a standard load and duration. All standards for this test are specified in SNI-06-2456-1996. The test method is as follows:

- Heating the sample with some of the temperatures in 120°C-150°C until it melts and then stirs until smooth
- Putting the sample into the mold until it dries
- Soak the sample in water at 25°C for 1 hour
- Perform a penetration test with a steel needle weighing 100gr for 5 seconds.

RESULT AND DISCUSSION

Product Thermal Behavior

Changes in the physical properties of multilayer plastics can be determined using Simultaneous Thermal Analysis (STA). The STA used in this test is a melting temperature measurement using Differential Scanning Calorimetry (DSC) [30]. The test was carried out by curing from 50ºC to 500ºC with a heat acceleration of 10K/min using the first heating (1st heating). Do not forget to adjust the nitrogen gas for the cooling stage at 20 ml/minute. These numbers are used as fixed variables so that the results can be uniform between one sample and another. In order to facilitate the research, a comparison curve was made between samples with the results of the DSC test. The curve can be seen in Fig. 3.

Based on the comparison of these curves, the plasma-treated sample has a melting temperature similar to that of multilayer plastic without plasma treatment which has a melting point of 163.8ºC [11]. A 30-second sample of plasma yields 163.7ºC at its melting point. Then at 60 seconds, the plasma sample has a melting point of 163.5ºC which is similar to multilayer plastic without treatment. It can be ascertained that the plasma treatment does not change the thermal properties of the multilayer plastic because there is no significant difference in degradation between samples. However, there is a crystallinity curve in the 155ºC region. This is due to several mixtures in the multilayer plastic composition. Although most of the composition is PP, it contains PE, HDPE, and printing inks that allow changing the sample graphics.

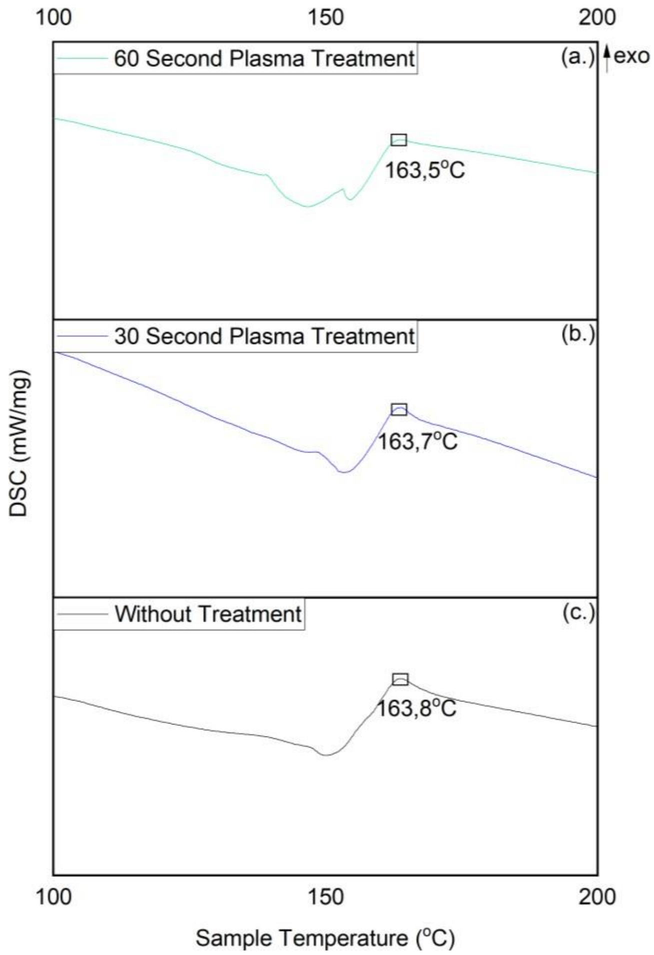

Figure 3. Multilayer Plastic DSC Curves (a) 60 Seconds, (b) 30 Seconds, and (c) Without Treatment

Product Wettability

Multilayer plastic has water repellent properties, which we usually know as hydrophobic. However, this makes the plastic unable to mix well with the bitumen, so plasma treatment is needed to change the nature of the multilayer plastic to become hydrophilic. To determine the wettability of the results of the treatment, it can be seen using the sessile drop test. The contact angle of the water droplets and ethylene glycol on the multilayer plastic can be determined through this test.

The longer the plasma treatment turns out affects the contact angle of the multilayer plastic. Plastic without treatment had a high contact angle of 71.92°, while plastic with plasma treatment for 15 seconds had an average contact angle of 50.94°. Multilayer plastic with plasma treatment for 30 seconds has an average contact angle of 42.97°, and this angle is smaller than plastic with plasma treatment for 15 seconds. For the latter, multilayer plastic with plasma treatment for 60 seconds has an average contact angle of 35.15°, and the angle is smaller than all previous treatments.

Using ethylene glycol, plastic without treatment has a high contact angle of 50.18°, while plastic with plasma treatment for 15 seconds has an average contact angle of 37.98°. Multilayer plastic with plasma treatment for 30 seconds has an average contact angle of 28.61°, and this angle is smaller than plastic with plasma treatment for 15 seconds. For the latter, multilayer plastic with plasma treatment for 60 seconds has an average contact angle of 24.52°, and the angle is smaller than all previous treatments. The surface tension can be found through the average of the contact angle test using water and ethylene glycol, which aims to determine how compatible the multilayer plastic is with bitumen. Bitumen samples were taken using the standard used in previous studies, namely 73.3° in water and 47.5° in ethylene glycol.

The critical surface tension of the liquid is used as the x-axis, where the water surface tension = 72 mN/m and the glycol surface tension = 48.5 mN/m. Cos (θ) as the y-axis, where the angle used comes from the angle of the contact angle test results on each liquid. A description of the average water and ethylene glycol from each treatment can be seen in Table 3.

Table 3. Average Table of Water and Ethylene Glycol from Each Treatment

Treatment	Water	Ethylene Glycol
Without Treatment	72,9	50,2
15 s Treatment	54,4	38
30 s Treatment	41,4	28,6
60 s Treatment	36,1	24,5
Bitumen	73,3	47,5

The data obtained from the contact angle analysis can be entered into the Zissmann equation to find the critical surface tension between the materials used, namely multilayer plastic and bitumen. The difference in surface tension between the materials will indicate the compatibility of the mixture between the two materials. The liquid compounds that will be used as a reference in this contact angle test are water and ethylene glycol with a surface tension of 72 mN/m and 48.5 mN/m, respectively. In making the Zissmann Equation Graph, the x-axis data represents the surface tension of the liquid compounds used, namely water and ethylene glycol. At the same time, the y-axis data is obtained from the results of cos (θ) from the corners of the contact angle test results of each dripping liquid. The critical surface tension can be found by entering the value of y = cos (0) = 1. This is because the critical surface tension is the surface tension that occurs when the solid surface is completely wetted (wetting angle = 0). Therefore, the critical surface tension is obtained as follows:
Critical surface tension (x) when y = cos(0) = 1

No treatment:
x = 24.102 mN/m

15 second treatment:
x = 24.193 mN/m

30 second treatment:
x = 26.278 mN/m

60 second treatment:
x = 28 mN/m

Bitumen:
x = 31.811 mN/m

After obtaining each x from each treatment, then compiled in the form of a comparison graph between the critical surface tension and time, as shown in Fig. 4. The graphical analysis shows that plasma treatment causes the compatibility between bitumen and multilayer plastic to increase, which can be seen from the decrease in the difference in surface tension between materials. Multilayer plastic with plasma treatment of 60 seconds has the smallest difference in interfacial tension with bitumen, namely: 31.811 – 28 = 3.811 mN/m, which shows the best compatibility compared to other experimental results.

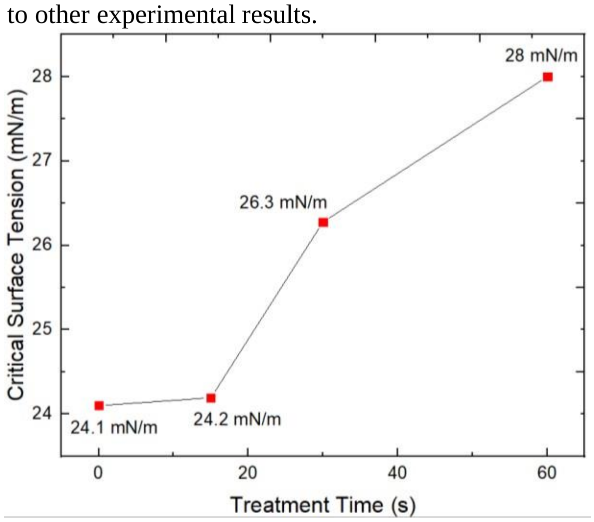

Figure 4. Comparison Graphic of Critical Surface Tension with Treatment Time

It can be concluded that the longer the plasma treatment, the plastic will have a smaller contact angle, which indicates a better wettability. However, based on additional experiments carried out for more than 60 seconds, the plastic cannot withstand the heat from the plasma, which causes the plastic to be scorched or charred and the surface to be perforated so that the plastic cannot be used again.

Experiments were carried out with samples 5 different times to show that the plasma treatment results were permanent. So that the plastic will not change its properties at any time, including after being mixed with bitumen, it is hoped that with the nature of the multilayer plastic, which tends to be more hydrophilic, it can mix well with the bitumen, and there will be no clumping between the plastics. This refutes several journals and claims that the results of plasma treatment only last for 30 minutes to 1 hour [27.28].

Product Compound Content

To find out whether there are differences between treated and untreated multilayer plastics, sample characterization was carried out using Fourier Transform Infrared Spectroscopy (FTIR). The results of the FTIR test function to determine the organic content contained in it by providing information in the form of peak absorbance bands or absorption of infrared waves at certain wavenumbers. Each has a different absorption capacity, so it will cause an increase in the amplitude of the vibrations of the atoms that are bound to each other. The offset spectra curve for the comparison between samples can be seen in Fig. 5.

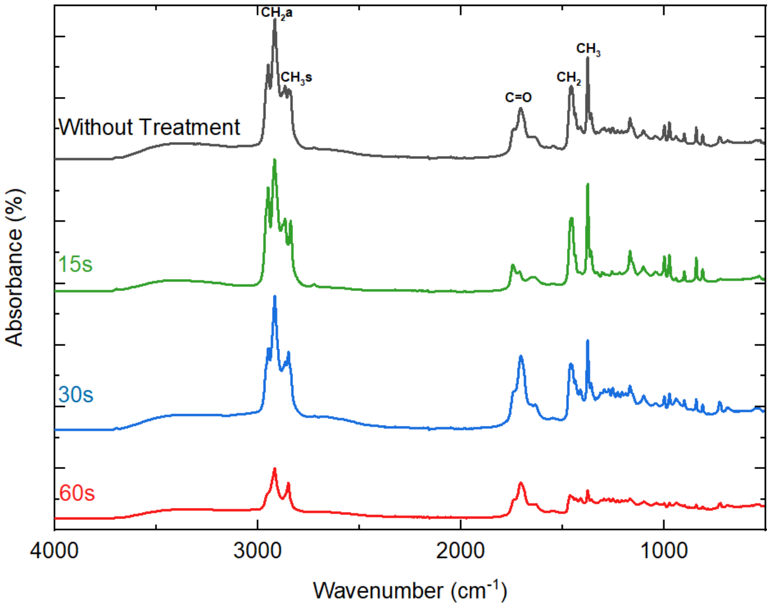

Figure 5. FTIR Spectra Curve Sample Offset Comparison

In Fig. 6, there is no significant degradation between samples. At first glance, the sample treated for 60 s has less absorbance at the 2917.29 cm^{-1} waves. This shows the low absorption capacity of the CH and CH_2 groups (aliphatic stretching group) due to the long plasma treatment on multilayer plastic which has the effect of scorching or scorching the instant noodle plastic wrap so that the bond disappears because the polymer chain is broken. Unlike the case with multilayer plastic which was given plasma treatment for 15 and 30 s, it had better absorbance than multilayer plastic without treatment. There is no significant degradation between samples. At first glance, the sample treated for 60 s has less absorbance at the 2917.29 cm^{-1} wave. This shows the low absorption capacity of the CH and CH_2 groups (aliphatic stretching group) due to the long plasma treatment on multilayer plastic which has the effect of scorching or scorching the instant noodle plastic wrap so that the bond disappears because the polymer chain is broken. Unlike the case with multilayer plastic which was given plasma treatment for 15 and 30 seconds (15s and 30s), it had better absorbance than multilayer plastic without treatment.

There is a slight difference in the wavenumber region around 1750 cm^{-1} and around 2800 cm^{-1}. In the 1750 cm^{-1} wavelength region, where the area shows a -C=O group, the difference between samples explains that the longer the plasma treatment, the more damage to the polymer chain due to plasma and the polymer chain radicals reacting with oxygen gas from the air, which is ionized due to plasma. The C=O structure is exposed to ozone and heat from the plasma, making the C chains that bind H and CH finally detach, and finally, the C compound is bound by the atmosphere. Thus, the C=O chain in the polypropylene breaks down [41].

Besides being degraded, multilayer plastic samples can also functionalize and create crosslinking between their groups. This is also influenced by CO, O_3, and OH produced by plasma and exposure to the air around the treatment. By forming functionalization and crosslinking, the polymer chain will be longer, and many new bonds will occur, so that the bond between the multilayer plastic and the bitumen will increase, making the modified bitumen harder stiffer. An illustration of the reaction mechanism can be seen in Scheme 2.

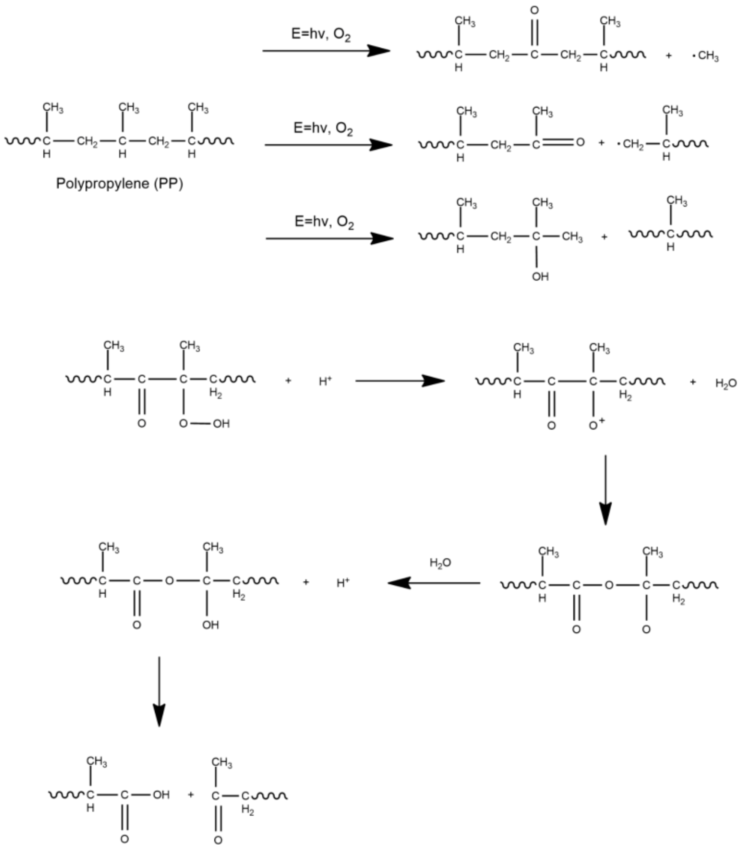

Figure 6. Illustration of Reaction Mechanism Due to Plasma [41]

Then at a wavenumber of 2800 cm^{-1}, which shows the CH and CH_2 groups (aliphatic stretching group). To analyze in more detail, it can be seen by comparing the peak ratio. The higher the percentage ratio, the higher the number of bonds in the peak. Samples from each treatment will be compared with samples without treatment. The peak that has been determined can be seen in Fig. 7.

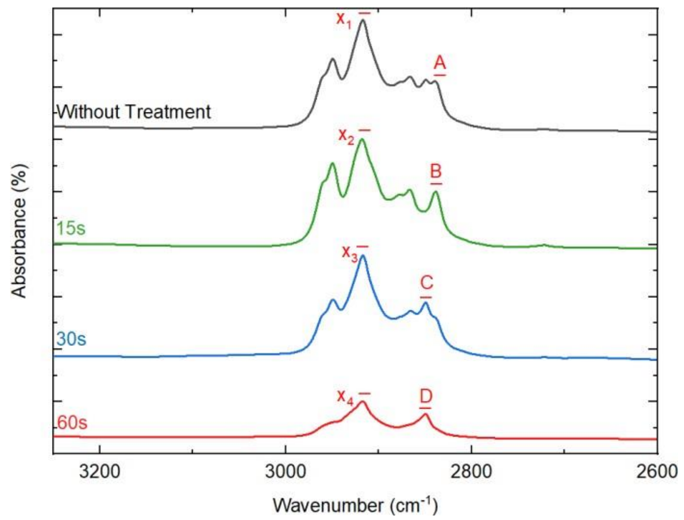

Figure 7. FTIR Spectra Curve at Peak CH2a and CH3s

From the results of this FTIR curve at the wavenumber distance of 2600 cm^{-1} to 3200 cm^{-1}, the peaks at 2900cm^{-1} and 2850cm^{-1} are taken to calculate the comparison. The value of X is used as the basis or backbone of each sample, and the peak of the CH group is used as a comparison.

After getting the absorbance number from each treatment, a curve can be formed to see the comparison line of the percentage of absorbance between plasma treatment samples guided by no treatment. With a simple equation, the results of the percentage ratio comparison are obtained, as shown in Fig. 8.

Through this curve, it can be seen that there is an increase in the percentage ratio of absorbance in all treatments. This indicates that there is an increase in group bonds in the plasma-treated samples. The O-H group of plasma provides functionalization of the CH_2a and CH_3s groups, as illustrated in Fig.5. However, the results of the sample with 60 seconds of treatment have a lower percentage ratio than 15 seconds and 30 seconds. This is due to the effect of excessive heat from the plasma, which causes degradation and O-H bonds in the atmosphere that attract C compounds from the CH_2a group or on the CH_3s.

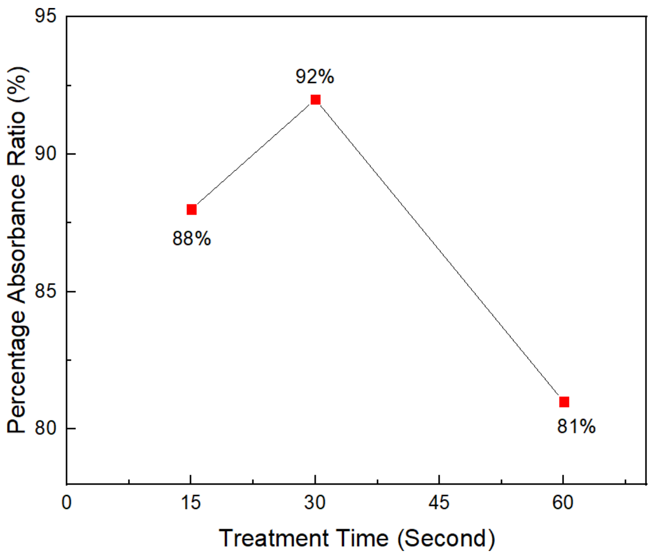

Figure 8. Comparison of Percentage Ratio of Peak Between Treatments

Analysis of Modified Bitumen

After all the results of the multilayer plastic testing have been compiled and analyzed, the next step is to analyze the results of mixing multilayer plastic with bitumen, namely modified bitumen. The best results from each sample can be determined through physical and chemical analysis of the sample. Physical analysis of the samples was carried out by testing penetration and ductility. The test was carried out to analyze the toughness and ductility of the modified bitumen and to match it with the standard requirements used by the Ministry of Public Works and Public Housing. Then to analyze the chemical properties of modified bitumen, an FTIR test was carried out. This test is used to ensure that there are additional groups indicating that there is a bond between the bitumen and the multilayer plastic

Penetration

Penetration testing is carried out to determine the hardness of bitumen. The penetration value is defined as the depth of the unit needle that enters the asphalt test object due to loading within a certain period of time, which is expressed in 0.1 mm. The load used, including the weight of the needle, is 100 grams, and the loading is carried out for 5 seconds at a temperature of 25ºC in 0.1 mm units. The higher the penetration value, the softer the sample should be. Along with the addition of plastic content, the penetration value will decrease, causing the modified bitumen to become harder [34-40]. Based on the standard requirements by the Ministry of Public Works and Public Housing, polymer asphalt has a penetration value of 40-80 [6]. The results of the penetration test can be seen in Fig. 9, and the sample code is based on Table 2.

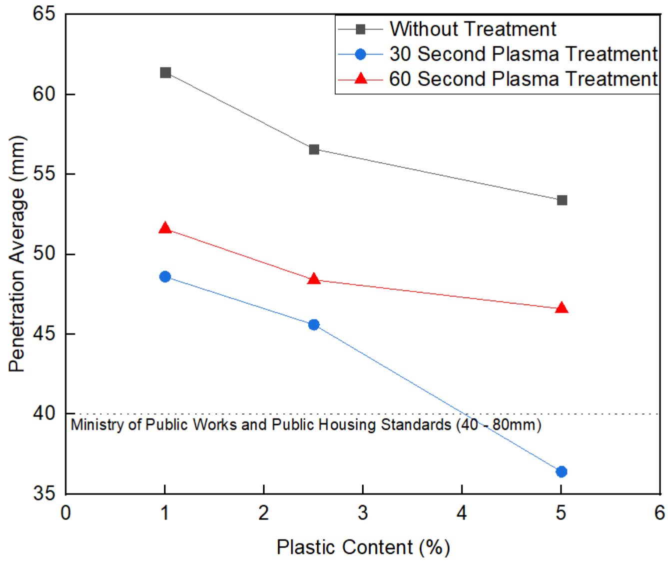

Figure 9. Penetration Check Results at 25ºC

Through the graph, it can be seen that there are optimal points in the bitumen mixture. Plastic content and duration of plasma treatment greatly affect the penetration results. The lower the penetration value, the harder the sample is indicated. According to the test results, the higher the multilayer plastic content in the bitumen, the harder the sample will be. The thing that makes the difference in the test results is the plasma treatment. The most optimal plasma treatment is for 30 seconds. This is shown because the average hardness in the sample has the lowest number compared to the others.

The BP_5T_{30} sample had the lowest average yield of 36.4 mm, followed by $BP_{2.5}T_{30}$ with an average of 45.6 mm. Both are samples with a plasma treatment time of 30 seconds. This can be assumed because there is a formation of the carboxyl group contained in the multilayer plastic, then bound to the aliphatic stretch (CH_2) group contained in the bitumen through a mixing process. Samples with a plasma treatment time of 60 s had higher penetration test results than those for 30 s because of the damage to several functional groups in multilayer plastic due to prolonged treatment at high enough temperatures which reduced the chain that binds between multilayer plastic and bitumen at high temperatures during mixing. This will also be explained in the FTIR test results. However, based on the standard requirements of the Ministry of Public Works and Public Housing, asphalt with a polymer mixture has a penetration value of 40-80 mm [6]. So that all treatments are still eligible to meet the requirements for the manufacture of polymer asphalt. However, with lower penetration, the application of aggregate in the manufacture of asphalt can be reduced. Thus, the cost of making asphalt will be cheaper.

Ductility

The asphalt ductility test is a test that can be used to determine the level of adhesiveness or ductility of hard asphalt. The higher the strength of the adhesion bond that occurs between bitumen and plastic, the better the compatibility, which will lead to good durability. The ductility test is carried out by measuring the longest distance drawn between the mold containing the sample before breaking at a specific temperature and tensile speed. The test was carried out at a temperature of 25°C and then withdrawn at 50 mm/min, where the sample was immersed in water. Based on the standard requirements by the Ministry of Public Works and Public Housing, polymer asphalt has a minimum ductility value of 40 cm [6]. The test results can be seen in Fig. 10, and the sample code is based on Table 2.

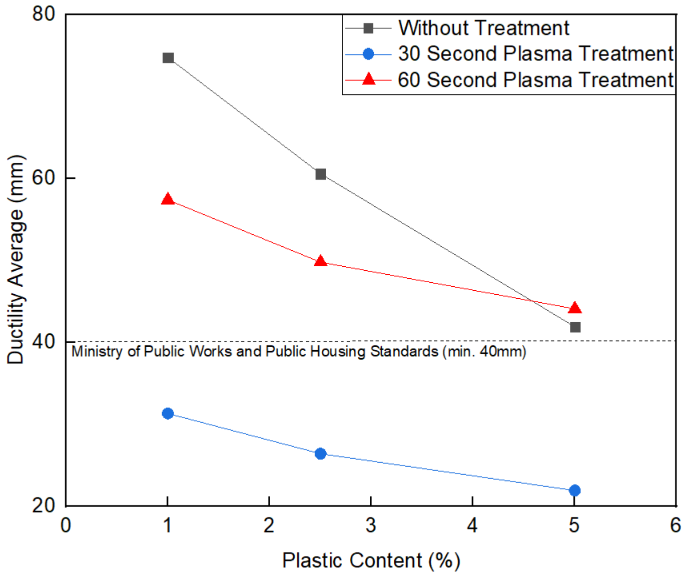

Figure 10. Ductility Test Results

Based on Fig. 10, the results of the ductility test also have the same pattern as the penetration test. There is an optimal composition in the test, namely the plasma treatment for 30 seconds. In the plasma treatment, it was seen that the modified bitumen sample had high stiffness. Same as penetration testing, which shows low penetration results. This has the same assumption as the previous test, which is estimated because there is a description of the carboxyl group contained in the multilayer plastic, which is then bound to the aliphatic stretch (CH_2) group where the group is present in the bitumen by going to through a mixing process. Samples with a plasma treatment time of 60 seconds have ductility test results that are more flexible than plasma treatment for 30 seconds because of the damage to several functional groups in multilayer plastic due to prolonged treatment at high enough temperatures which reduces the chain that binds between multilayer plastic and bitumen at high temperatures during mixing. However, based on the standard requirements of the Ministry of Public Works and Public Housing, asphalt with a polymer mixture has a minimum value requirement of 40 mm so that the results that allow it to be used and have excellent ductility and hardness are modified bitumen with multilayer plastic which is given plasma treatment for 60 s [6].

CONCLUSIONS

After doing all the research series, starting from sample making, testing, and overall product analysis, it can be concluded that the cold plasma treatment can change the physical properties of the multilayer plastic surface from hydrophobic to hydrophilic due to the formation of -COOH, C=O, and -OH functional groups on the multilayer plastic surface. This is due to the plasma treatment under 60 seconds does not have a side effect of degradation of multilayer plastic. Therefore the optimal cold plasma treatment time is 60 seconds because at that point, the plastic has the best wettability, and the plastic is not damaged by excessive heat, and multilayer plastic adds hardness to the bitumen. Thus, the plasma treatment for 60 seconds with a plastic composition of 5%.

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