PERFORMANCE EVALUATION OF PRE-VULCANIZED LIQUID NATURAL RUBBER (PVLNR) IN HOT MIX ASPHALTIC CONCRETE

*Irfan1,2, Bambang Sugeng Subagio3, Eri Susanto Hariyadi4 and Indra Maha5

1,3,4 Faculty of Civil and Environmental Engineering, Bandung Institute of Technology, Indonesia; 2 Faculty of Civil Engineering, Teuku Umar University, Indonesia; 5 Bina Marga Office of West Java, Indonesia

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ABSTRACT: Liquid natural rubber as an asphalt modification material to replace polymer, to improve the rheology of asphalt, increase resilient modulus mix, increase the resistance of high-temperature mixtures but remain flexible at low temperatures. Pre-Vulcanized Liquid Natural Rubber (PVLNR) modification of conventional liquid natural rubber is used explicitly as an asphalt modification material. With the limited research on the use of PVLNR, it is necessary to test for the performance analysis of PVLNR modified asphalt. This study aims to analyze the effect of adding PVLNR modified asphalt on asphalt rheology and mix performance. This laboratory-scale experimental research includes rheology testing of modified asphalt, mix performance in marshall testing, and resilient modulus. Based on the test results, the addition of PVLNR 3%, 5%, and 7% (of the asphalt weight) could decrease the penetration value, increase the viscosity and melting point, and provide a better elastic recovery of 55%, 57%, and 60%. Volumetric parameter analysis, such as density PVLNR modified Hot Mix Asphalt (HMA), tends to be equivalent to the HMA control mixture. Resilient modulus in samples using gyratory compactor increased with an increasing percentage of PVLNR. At a temperature test of 41 ºC, the resilient modulus of PVLNR modified HMA was higher with a ratio of 1.7, 1.8, and 2.2 compared to the HMA control. PVLNR can substitute asphalt to save asphalt consumption by 0.17%, 0.28%, and 0.39%, respectively, of the asphalt weight.

Keywords: Pre-Vulcanized Liquid Natural Rubber, Resilient Modulus, Elastic Recovery.

1. INTRODUCTION

With a production capacity of 3 million tonnes per year, Indonesia is one of the largest rubber producers. The decline in rubber prices in the international market has prompted the government to increase domestic rubber consumption, such as encouraging rubber as a modified asphalt material [1]. As a polymer replacement, liquid natural rubber improves asphalt pavement performance and resistance to rutting at high temperatures [2-5].

With the high ammonia content to cause sparks and release excess ammonia gas during the heating process, there are a few problems with conventional liquid natural rubber in the field. Moreover, its unstable nature also causes agglomeration of rubber particles during the modification of the process [6].

Research and Development Center for the Implementation of Road and Bridge Technology collaborates with Indonesia Rubber Research Institute in Bogor to develop rubber asphalt technology consisting of liquid natural rubber, SIR solid rubber, and crumb rubber. Lastly, a modification of liquid natural rubber into Pre-Vulcanization Liquid Natural Rubber (PVLNR).

Producing Pre-Vulcanized liquid natural rubber (PVLNR) by blending conventional natural rubber with dispersing chemicals consisting of activators, accelerators, antioxidants, and sulfur, and stirring for 4 hours at 70ºC was carried out [7].

PVLNR as modified asphalt is better than conventional liquid natural rubber has a lower viscosity, reducing the mixing temperature by ±10 ºC [1]. PVLNR has a high cohesion power, so elastic recovery is better than conventional liquid natural rubber [6,8,9].

PVLNR modified emulsion asphalt can increase thermal stability and increase the characteristics of PVLNR modified emulsion asphalt. The swelling ratio affects the interaction (cross-link) between rubber and asphalt of the vulcanization process determines the swelling ratio of PVLNR. The Swelling ratio of 7 to 15 for the vulcanization process for 4 hours. The PVLNR is more stable and can store for up to 300 days from the vulcanization process [10,9].

Studies on using asphalt modified liquid natural rubber are still limited [11] and become more limited for PVLNR rubber types because polymer modified asphalt is more widely known. In addition to the limited availability of natural
rubber raw materials only in countries that produce natural rubber. The study of the use of PVLNR as an asphalt modification still leaves drawbacks, especially for rheology modified asphalt, volumetric properties, and resilient modulus mix. The mixing method, such as mixer speed, temperature, and length of the modification process, is strongly influenced the performance of liquid natural rubber asphalt modification [12,13,14].

This study aims to observe the effect of the addition of PVLNR to Asphalt Concrete - Binder Course (AC-BC) mixtures. The impact on the value of rheology of the modified asphalt and its resilient modulus will be. PVLNR addition of 3%, 5% and, 7% by weight, will be used in the sample preparation.

2. MATERIALS PREPARATION

2.1 Aggregate

Andesite type aggregate material used in hot asphalt mixture came from crusher stone PT Kadi International, Karawang Regency, West Java Province. Table 1 showed the aggregate properties and aggregate gradation, respectively.

Table 1. Aggregate properties

| Test Method | Aggregate |
|-------------|-----------|
| Test        | Coarse    | Fine     |
| Specific Gravity | ASTM C127-84 | 2.619 | 2.656 |
| Water Absortion (%) | ASTM C127-84 | 1.763 | 0.732 |
| Los Angles Abrasion (%) | ASTM C131-76 | 5.12  | -     |
| Flakiness and Elongation (%) | ASTM D-4791  | 7.2   | -     |

The gradation used referred to the provisions of the Bina Marga General Specifications of Roads and Bridges 2018, with Asphalt Concrete - Binder Course (AC-BC) gradation type. The gradation design used the middle boundary in the Fuller curve [11], as shown in Fig. 1.

2.2 Pre-Vulcanized Liquid Natural Rubber

PVLNR (Pre-vulcanized Liquid Natural Rubber) is a product from Indonesia Rubber Institute in Bogor, was explicitly developed to be used as an asphalt modification material. The PVLNR has better quality from the initial analysis. The type of rubber specifications that is good for use on asphalt mixtures has not been determined. The properties of the PVLNR can be seen in Table 2.

Table 2. Properties PVLNR

| Properties            | Test Method | Result |
|-----------------------|-------------|--------|
| Total Solid Content, % | ASTM D 1076-15 | 58.53  |
| Mechanical Stability, (Second) | ASTM D 1076-15 | 900    |
| pH                    | ASTM D 1076-15 | 10.99  |
| Chloroform Number     | LP-PPK      | 4      |
| Acetone Content, %    | ASTM D 297-15 | 8.19   |
| Polymer Content, %    | LP-PPK      | 88.07  |

2.3 Asphalt Pen 60/70

The asphalt used was oil asphalt pen 60/70 produced by PT Pertamina (Persero). This type of asphalt commonly used as a road pavement material in Indonesia, with basic properties described in Table 3.
Table 3. Basic Properties of asphalt pen 60/70

| Properties                  | Value | Specifications |
|-----------------------------|-------|----------------|
| Penetration (0.1 mm)        | 63.2  | 60-70          |
| Softening Point (°C)        | 49.65 | ≥ 48           |
| Ductility (cm)              | ≥ 100 | ≥ 100          |
| Flash Point (°C)            | 248   | ≥ 232          |
| Solubility (%)              | 99.92 | ≥ 99           |
| Specific gravity            | 1.04  | ≥ 1.0          |
| Weight Loss TFOT (%)        | 0.003 | ≤ 0.8          |
| Penetration TFOT (%)        | 80.4  | ≥ 54           |
| Ductility TFOT (cm)         | ≥ 100 | ≥ 50           |

2.4 Asphalt Binder Modification

PVLNR modified asphalt produced with wet mix method by mixing PVLNR of 3%, 5%, and 7% (of the asphalt weight) using a mixer at a temperature of 150 °C for 20 minutes with a speed of 2000 rpm. PVLNR was added slowly to the hot asphalt, gradually to reduce foam's appearance due to water evaporation [12,13,14].

2.5 Asphalt Mix Performance Test

The experimental mixture performance test conducted using a marshall tool to analyze the mixture performance, volumetric analysis, then continued with fundamental tests using the universal testing machine (UTM) with a gyratory compactor.

3. TEST RESULT AND DISCUSSION

3.1 PVLNR Modified Asphalt

Asphalt modification with PVLNR carried out in the ITB Road and Traffic Engineering laboratory. The asphalt modification of glory tested according to, the Laston Interim Special Specifications with Natural Rubber Content (Skh-1.6.25) and the General Specifications of Bina Marga 2018 [14,11]. The test result is shown in Table 4.

The higher the percentage of PVLNR modified asphalt, the penetration will decrease, and the melting point will be higher, and elastic recovery increased by 55%, 57%, and 60%. It was caused by the presence of rubber particles that enter the modified asphalt.

The entry of rubber particles occurs through a heating process that causes the asphalt to expand and melt so that these rubber particles can enter the asphalt. These rubber particles then absorb solvents, which are lubricating liquids from asphalt. The amount of absorption depends on the surface area of liquid natural rubber particles. Although the asphalt is getting harder, the elastic recovery continues to increase. The size of PVLNR particles is smaller, 50 nm [15], density 0.970 g/cm³ and 0.985 g/cm³ [10]. The bonding process between these rubber particles and asphalt contributes to improving the elastic recovery of modified asphalt; with smaller particle sizes of 50 nm, these rubber particles can function as nano binders [10]. Increased elastic recovery is affected by the presence of rubber particles that absorb asphalt (solvent) so that these rubber particles swell with a swelling ratio from 7 to 15 [16].

An increase in the content of liquid natural rubber increases the PG grade of asphalt. The PG values of LNR0, LNR3, LNR5, LNR7 liquid natural rubber modified asphalt, according to AASTHO M320 specifications, become PG64-10, PG64-16, PG70-16, and PG70-16 [15]. This related to the asphalt resistance to an increase in temperature [17].

Table 4. Basic PVLNR Modified Asphalt [9]

| Properties                  | PVLNR 3% | PVLNR 5% | PVLNR 7% | Specifications |
|-----------------------------|----------|----------|----------|----------------|
| Penetration (0.1 mm)        | 60.8     | 57.8     | 52.7     | Min. 50        |
| Softening Point (°C)        | 51.8     | 52.9     | 53.65    | ≥ 52           |
| Ductility @ 25 °C (cm)      | ≥ 100    | ≥ 100    | ≥ 100    | ≥ 100          |
| Flash Point (°C)            | 323      | 325      | 328      | ≥ 232          |
| Solubility in C₂HCl₃ (%)    | 99.72    | 99.58    | 99.42    | ≥ 99           |
| Specific Gravity            | 1.033    | 1.033    | 1.033    | ≥ 1.0          |
| Weight Loss TFOT (%)        | 0.000    | 0.000    | 0.000    | ≤ 0.8          |
| Penetration TFOT (%)        | 85.3     | 86.6     | 87.1     | ≥ 54           |
| Ductility TFOT (cm)         | ≥ 100    | ≥ 100    | ≥ 100    | ≥ 50           |
| Elastic Recovery (%)        | 55       | 57       | 60       | ≥ 30           |
The content of liquid natural rubber in asphalt changes the chemical composition of asphalt. There are contents of Asphaltene and Nitrogen base, increased content of Asphaltene, which is a polar compound that is insoluble in n-heptane, improves the rheological properties of asphalt [18]. Asphaltene content has more effect on elasticity modulus than viscosity modulus, which increases resistance to the rutting factor \((G^*/\sin \delta)\), which is also higher compared to the fatigue crack factor \((G^*\sin \delta)\). Besides, an increase in asphaltene content is also more influential on long-term ageing (PAV) than on short-term ageing (RFTO) [19]. Nitrogen base is a compound that is soluble in n-heptane. The Nitrogen base's content functions as an anti-stripping agent, which increases the adhesion of asphalt, but the content of the Nitrogen base has high reactivity, so it is easy to undergo ageing [20].

### 3.2 Mixing and Compaction Temperatures

The mixing and compaction temperature is determined from the viscosity value using the Sybold Furrol tool, for based on the general specifications of Bina Marga 2018, the mixing temperature is determined to be \(170 \pm 20\) cSt, and the compaction temperature is \(280 \pm 30\) cSt, the test results are in Table 5.

| Asphalt Type      | Temperature (°C) |
|-------------------|------------------|
|                   | Mixing | Compaction |
| Asphalt Pen 60/70 | 155   | 145        |
| PVLRN 3% modified | 162   | 152        |
| PVLRN 5% modified | 166   | 155        |
| PVLRN 7% modified | 175   | 165        |

Increasing the percentage of PVLRN in the asphalt binder, increasing the asphalt binder's viscosity so that the mixing and compaction temperature increases up to \(20^\circ\) for the PVLRN percentage of 7%, as shown in Table 5. In previous research for conventional liquid natural rubber modified asphalt, the maximum percentage of LNR in asphalt is 7%, with a viscosity value of <3 Pa.s [15].

### 3.3 Marshall Test

The estimated range of asphalt content use was obtained from calculating planned asphalt content (Pb), which is 5.25%.

The results of marshal testing on the optimum binder content (OBC) as seen in Table 6.

Optimum binder content (OBC) for the mixture increases with the increase in the percentage of PVLRN in the mix. Caused the asphalt that is getting harder, so it takes more percentage of asphalt to cover the aggregate properly. The use of pure asphalt decreases with an increase in the percentage of PVLRN; this is because PVLRN substitutes asphalt usage, resulting in savings in asphalt consumption of...
0.17%, 0.28%, and 0.4% (of the percentage OBC) for each modified asphalt mixture of HMA PVLNR3, HMA PVLNR5, and HMA PVLNR7.

The stability of the mixture increases with the addition of the percentage of PVLNR, as shown in the figure. 2. The rheology of asphalt influenced the increased stability value of the mixture. Rubber particles increase the adhesion of asphalt and the cohesion between asphalt and aggregate; this property makes the mixture stiffer so that the mix's stability increases [21]. Higher stability value a polymer-modified asphalt mixture as an indicator that the mix can take traffic load without experiencing permanent deformation and better flow as an indicator that the mixture is resistant to plastic deformation [5,21,22].

The mixture's density value decreases with the addition of asphalt percentage, except for the percentage of void in the mix (VIM) that increases, as shown in the figure. 3. Enhancement viscous properties of asphalt modified influence the increase in density. This bitumen's viscosity is affected by temperature. A good density can achieve under the right viscosity and compaction energy conditions [9]. The increase in VIM value is because the asphalt can cover the aggregate well, which is influenced by increased adhesion and cohesion properties of modified asphalt [1,9].

3.4 Resilient Modulus Test

Resilient modulus is the asphalt mixture's ability to take loads under conditions that remain elastic [24]. The resilient modulus test using UTM (universal testing machine) at the ITB Highway and Traffic Engineering Laboratory refers to the ASTM D4123 Standard Test Method for Indirect Tension Test for Resilient Modulus of Bituminous Mixtures [25]. Test conditions set at a loading pulse width of 250 ms and a pulse repetition period of 3000 ms. The test conducted at temperatures of 20 °C, 30°C, 41°C, and 50 °C.

According to the Indonesia Road Pavement Design Manual 2017 [26], asphalt pavement temperature can express as an annual weighted mean asphalt pavement temperature (WMAPT). For the Indonesian climate, WMAPT ranges from 38°C (for mountainous areas) to 42°C (coastal areas). The resilient modulus of the asphalt mixture used in the design chart was calculated based on the WMAPT assumption of 41 °C [11].

The percentage of asphalt used was the percentage of OBC from the Marshall test results, which 5.5%, 5.58%, 5.68%, and 5.73% consecutively. The compaction using the gyratory tool with a control with a Marshall density test result. Compaction of the sample mix resilient modulus test using a gyratory compactor (SGC), which refers to the standard AASTHO T 312 Preparing Hot-Mix Asphalt (HMA) Specimens utilizing the Superpave Gyratory Compactor [27].
The mixed resilient modulus increases with increasing percent PVLNR, as shown in Fig. 7, Fig. 8, Fig. 9, Fig. 10. The good distribution of rubber particles in the asphalt is affected by increased resilient modulus value; these rubber particles are highly reactive, influencing the adhesion properties of asphalt and increasing compatibility with aggregates [2, 25].

The content of PVLNR particles enhances bitumen rheology, where the asphalt becomes harder. A certain amount extent, the increase in the hardness of the asphalt is related to the stiffness modulus of the asphalt; the asphalt stiffness modulus is related to the resilient modulus mix, the higher the stiffness modulus of the asphalt, the higher the resilient modulus of the mixture [28, 29].

The resilient modulus of the PVLNR modified asphalt mixture is also better in high temperatures compared the HMA control, as shown in Fig. 9, Fig. 10. Resilient modulus increase due to the addition of PVLNR increases the asphalt softening point temperature so that the mixture is more resistant to high temperatures [15]. In previous research, the use of epoxidized natural rubber as an asphalt modification increases the mix's resilient modulus, especially at high temperatures [30]. The high value of this modulus is inversely proportional to the declining density and higher VIM (void in mix) so that the asphalt performance affects the modulus of the mixture [30]. The higher the test temperature, the modulus decreases. The HMA PVLNR has a better condition at a temperature of 41°C. Based on the 2017 Pavement Design Manual, the mixture modulus for the AC-BC HMA layer is 1200 MPa [26].
The increase in the percentage of PVLNR has a negative effect, the asphalt gets harder, more viscous, and softening points increase, increasing the temperature of the mixture production.

The quality of PVLNR modified asphalt highly influences modification processes such as mixing temperature and mixer speed. Segregation, such as rubber lumps on the asphalt, is caused by the incomplete modification process. This initial study is still limited; therefore, it is necessary to carry out further studies to observe modified asphalt's degradation properties with time and improve the asphalt modification method.

4. CONCLUSION

PVLNR can be used as a modified asphalt material, especially for rubber-producing countries where sufficient raw material is available. From the results of the study, several conclusions can be drawn:
1. The percentage of PVLNR in the asphalt decreases penetration and increases the softening point. This is due to the rubber particles that fill between the asphalt parts and absorb the asphalt content (solvent), so that the asphalt becomes harder.
2. The percentage of PVLNR in asphalt increases the elastic recovery value affected by the bonding process between rubber particles and asphalt, increasing the elastic recovery from modified asphalt.
3. The OBC of the mixture increases with an increasing percentage of PVLNR. This is due to the increasing number of rubber particles in modified asphalt, which absorbs asphalt (solvent).
4. The use of PVLNR saves asphalt use, respectively 0.17%, 0.28, and 0.4% (of the OBC percentage).
5. The resilient modulus increases with an increasing percentage of PVLNR. It has good performance, especially for high temperatures. This is influenced by the content of asphalt well dispersed PVLNR particles, increasing the bonding of adhesion and the cohesion of asphalt and aggregates.

5. ACKNOWLEDGEMENT

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