Effect of depolymerized block skim rubber (BSR) on the physical properties of modified asphalt

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ABSTRACT
The asphalt pavement susceptibility was influenced by traffic load and temperature. Therefore, modified asphalt has been done to improve the asphalt performance. Depolymerized block skim rubber (BSR) was used as asphalt modifier. BSR is a low quality crumb rubber, which made from skim (byproduct of creamed latex). This investigation aims to determine effect of depolymerization of BSR on the physical performance of modified asphalt. Initially, BSR was depolymerized by mechanical action (mastication) with different time of 8, 16, and 24 min. Then, the asphalt modifier, depolymerized BSR was added to asphalt with different ratio 3, 5, 7%. The softening point and mixing time of asphalt were compared with modified asphalt. The performance of modified asphalt showed that asphalt modifiers increased the softening point and mixing time of asphalt. Maximum softening point reached 54.30°C by 7% BSR (16 min. of mastication time). The modified asphalt had 25.70°C softening point higher than asphalt. It was conclude that depolymerized BSR can increase the performance of asphalt (softening point), although increase the mixing time of it. Moreover, it could give alternative to reduce cost of modified asphalt pavement making.

Keywords: Asphalt, block skim rubber, depolymerized, modified asphalt

INTRODUCTION
Asphalt pavement is a transportation facility on this modern era. Asphalt pavement has high frictional force, so the vehicle tires are not slip easily on wet or dry condition. However, asphalt pavement susceptibility was influenced by traffic load and temperature. Asphalt becomes brittle and has low-thermal cracked at low temperature (0-30°C), whereas it flows and suffers from rutting at high temperature (40-65°C) (Lewandowski, 1994). In tropical country, the highest temperature in summer time can attain 50°C, which is higher than the softening point of asphalt (± 48°C). This will reduce the service life of the pavement.

The potential solution for addressing today highways problems is mixing a polymer with the asphalt. The studies of polymer modified asphalt showed that the overall polymer mixture on asphalt had increase the performance of asphalt (Ali et al., 2013; Bakar et al., 2018; Ali et al., 2015; Huang et al., 2017). The majority polymer used for asphalt modifier were styrene–butadiene–styrene (SBS) (Chen et al., 2002; Martinez-Estrada et al., 2010; Kök & Colak, 2011; Liu et al., 2014) and technically specified rubber (TSR) as crumb rubber or tire scrap form (Bai et al., 2016; Presti, 2013; Kök & Colak, 2011; Prastanto, 2014).

Unfortunately, SBS, as synthetic rubber, and TSR, as natural rubber, are expensive materials compared with asphalt and the substantial improvement in performance could only achieve at an appraisal 25% increase on material costs. Besides that, Indonesia has to import synthetic rubber, meanwhile Indonesian TSR (SIR 20) has been using as raw material of high quality products, such as tire, dock fender, bridge bearing, etc. One of the alternative solution is to utilize the second grade of natural rubber, which is block skim rubber (BSR).

BSR is a low quality of crumb rubber, which is made from skim. Skim natural rubber latex is the side product of centrifugal concentration of
natural rubber latex containing high protein (George et al., 2009). Skin contains 4-8% dry rubber content (DRC) (Blackely, 1966; George et al., 2009). Recently, the assumption of skin production per year in Indonesia reached 2,549 ton (Haris et al., 2010). The price of BSR per kg is around 40% of the SIR 20 price (7,743 IDR). It was cheaper than the price of SIR 20 per kg (19,358 IDR) and SBS per kg (182,108 IDR) on August 2018. Utilizing this waste material might turn out the solution of a waste disposal-problem and construction cost of roads.

BSR, as natural rubber, has long polymer molecules chain that can lengthen the mixing time and needs higher energy to mix the asphalt. Depolymerization is polymer decipherment to shorten this polymer molecules chain. Depolymerization can be held by chemical (Prasoetsopha et al., 2011), thermal and mechanical (Nair et al., 1995) or mechanical action (Prastanto, 2014) on latex phase (Prasoetsopha et al., 2011; Nair et al., 1995) or rubber phase (Prastanto, 2014). In this research, depolymerization held by mechanical action using open mill at three different times of mastication.

Prastanto (2014) reported that mechanical depolymerization during 24 minutes decreased the molecular weight refer to decreasing of Mooney viscosity. Then, the depolymerized BSR will be used to modify asphalt making. This investigation aims to investigate the effect of depolymerized BSR on the physical performance of modified asphalt and to find optimum composition of depolymerized BSR as modifier in modified asphalt.

**MATERIALS AND METHODS**

**Material and Sample Preparation**

Asphalt (P60), obtained from Binamarga, was used as bitumen for mixture preparation. The skim rubber was supplied by PTPN 8. Afterward, the skim rubber was coagulated with sulfuric acid 2% and dried with oven. The properties of BSR was analyzed by standard test of Indonesian TSR (SIR 20). The properties of BSR are listed in Table 1. This properties was compared with quality standard of SIR 20.

BSR was depolymerized by mechanical action using open mill. The temperature of mastication was controlled under 60°C. Depolymerization was held on two period gradually. Firstly, BSR (200 g) was masticated for 5 minutes then added with 0.2 phr (per hundred rubber) peptizer. Furthermore, BSR was masticated again for three different time of mixing (8, 6, 24 min.). Then, 0.6 phr hydroxylamine neutral sulfate (HNS) was added into BSR. The depolymerized BSR per each mixing time was utilized as asphalt modifier.

**Polymer Modified Asphalt Mixture**

A mixture of depolymerized BSR and asphalt was held on high temperature design. Asphalt (300 g) was added on stainless flask and heated by electrical stove at 160°C. Sample temperature and heating rate was controlled by thermocouple. Mechanical rotor was started once the asphalt homogenous melt, rotor speed was adjusted to 20 rpm. Furthermore, depolymerized BSR was resized into small pieces sample prior to mix with liquid asphalt. The small pieces depolymerized BSR per each mixing time was added 3, 5, 7% (% wt) into liquid asphalt separately. This mixture was mixed until homogenous. Then, the physical properties of polymer modified asphalt was analyzed.

**Testing Method**

**The properties of depolymerized BSR**

An analysis of depolymerized BSR properties was held to determine the effect of depolymerization on BSR properties, especially the polymer molecular chain length. The properties of depolymerized BSR was analyzed by standard test of SIR 20. The parameter properties analyzed including:

a) Initial plasticity (Po) (SNI 06-1903-2000)

15-25 g samples was prepared previously. A sample was milled by open mill to homogenize the sample and flatten it became 1.6-1.8 mm thickness. Then, the sheet of sample was folded to increase the sample thickness became 3.2-3.6 mm. A sample was cut down into three round shapes using Wallace punch. Furthermore, a round shape sample was put between two sheets of cigarette paper (TST) and tested it using Wallace plasticity meter. The measurement of initial plasticity was held twice to obtain accurate data.

b) Plasticity retention index (PRI) (SNI 06-1903-2000)

15-25 g samples was prepared previously. A sample was milled by open mill to homogenize the sample and flatten it became 1.6-1.8 mm thickness. Then, the sheet of sample was folded to increase the sample thickness became 3.2-3.6 mm. A sample was cut down become three round
shapes using Wallace punch. Furthermore, a round shape sample was put in oven at 140±0.2°C for 30 minutes. The sample plasticity after aging was measured using Wallace plasticity meter as Pa. The measurement of plasticity retention index was held twice to obtain accurate data. PRI value was calculated with this formula:

\[ \text{PRI} \text{ (%) = } \frac{\text{Pa}}{\text{P}_0} \times 100 \]

\( \text{P}_0 \) = Initial plasticity  
\( \text{Pa} = \text{Plasticity after ageing in oven at 140 ± 0.2°C for 30 minutes} \)

c) Mooney viscosity (ASTM D-1076-97)

Initially, the temperature of Mooney viscometer was set 100°C for 1 minute. After that, a sample (± 25 g) was put on the up and down the rotor in that machine and the rotor was rotated for 4 min. The energy required to rotate the rotor was read on the scale. The unit of Mooney viscosity value is M(1’+4’)/L100°C.

The physical properties of polymer modified asphalt

The properties of the polymer modified asphalt was analyzed to determine the effect of addition asphalt modifier on asphalt, compared to asphalt. The physical properties of modified asphalt was analyzed including:

a) The mixing time of modified asphalt

The mixing time of modified asphalt is the time needed to blend the asphalt and polymer. The calculation of mixing time started when asphalt melted until all of the polymer mixture homogenous with asphalt. The calculation time measured by stop watch.

b) Softening point

Softening points were used to determine the temperature at which a phase change occurs in the binder. Initially, two brass rings was layered with glycerin, then chilled at room temperature for 30 min. A sample was melted to become liquid phase, then the sample was poured into brass ring. That sample was chilled at room temperature for 1 h. After that, the brass rings was fitted with ball centering guide (put into the center of brass rings) inside ring holder. Furthermore, the tools measurement was put into container of aqua. In other side, the thermometer was put into the hole of tools measurement (Figure 1). The container was heated so that its temperature rise reached 5°C/ min. The softening point was taken as the temperature at which the sample became soft enough to allow the ball, enveloped in the sample material, to fall a distance of 25.4 mm. This was recorded as the softening temperature.

RESULTS AND DISCUSSION

The Properties of Depolymerized BSR

The properties of BSR (Table 1) shows that BSR has quietly met a SIR standard, which are ash content and dirt content properties. That result describes that BSR has low ash and dirt content. The skin latex has low dirt content and it potentially becomes high quality rubber if the protein content can be increased and problems arising from coagulation are solved (Alex & Nah, 2006). Nevertheless, BSR has high nitrogen and volatile materials content (such as water vapor or serum) and has shorter polymer molecular chain and lower stability on high temperature than SIR 20.

Both \( P_0 \) and Mooney viscosity are decreasing with the increasing of mastication time (Figure 1). Depolymerization held by milling process using open mill. Masticating rubber for longer time, will make rubber becomes more plastic than the unmasticated. An elastic rubber has low molecular chain length and molecular weight. The addition of peptizer on mastication process has gave polymer molecule weight decrease effect too on depolymerized BSR.

The depolymerization process comparison of BSR and SIR 20 (Figure 2) shows insignificantly different. The raw material of polymer does not influence the succession of depolymerized process. The depolymerization at 24 minutes of mastication time has the highest percentage of Mooney viscosity degradation on both of that rubber. The BSR depolymerized can achieve 93.54% degradation of Mooney viscosity, whereas SIR 20 can achieve 86.46% degradation of Mooney viscosity. It means that the depolymerization process of BSR is more successful than SIR 20, even though SIR 20 has better properties than BSR.
The Physical Properties of Polymer Modified Asphalt

Asphalt is a heterogeneous material consists of immiscible phase of asphaltenes and maltenes. Asphaltenes are polyatomic hydrocarbons of relatively high molecular weight. Maltenes are a mixture of resins, waxes, aliphatic and aromatic compounds (Browarzik et al., 1999; Murgich et al., 1996). The addition of polymer in asphalt could raise the properties of asphalt (Kanitpong & Bahia, 2005; Chen et al., 2002; Hussein et al., 2006; Tuntiworawit et al., 2005). The morphology of asphalt and polymer blend was studied using SEM showed that both of them was blended homogenously (Chen et al., 2002; Xiang et al., 2009). This result concluded that polymer had compatible with asphalt matrix, which could increase the asphalt performance.

The mixing and applying technique of asphalt-rubber to roads mainly use two ways, the dry and wet process (Huang et al., 2007). Crumb rubber is used as a part of the aggregate in the hot mixture to replace some of the solid fraction in a dry process (Azizian et al., 2003). While others, crumb rubber is added to the asphalt cement mixture in a wet process. In this research, the mixture of asphalt and depolymerized BSR was prepared by wet process. The wet process can rectify the rutting resistance, resilience modulus, and fatigue cracking resistance of asphalt mixtures (Mashaan et al., 2014) because wet process stimulates rubber particles absorb the aromatic fractions of asphalt as much as possible, and increases the viscosity of the asphalt (Ibrahim et al., 2013).

Table 1. BSR properties.

<table>
<thead>
<tr>
<th>Properties (%)</th>
<th>BSR</th>
<th>SIR 20</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volatile matter content</td>
<td>2.78</td>
<td>Max. 0.8</td>
</tr>
<tr>
<td>Ash content</td>
<td>0.62</td>
<td>Max. 1.0</td>
</tr>
<tr>
<td>Dirt content</td>
<td>0.013</td>
<td>Max. 0.2</td>
</tr>
<tr>
<td>Initial plasticity (P₀)</td>
<td>26.00 Min. 30</td>
<td></td>
</tr>
<tr>
<td>Plasticity retention index (PRI)</td>
<td>15.40 Min. 50</td>
<td></td>
</tr>
<tr>
<td>Nitrogen content</td>
<td>2.23</td>
<td>Max. 0.6</td>
</tr>
</tbody>
</table>

Table 2. The properties of depolymerized BSR.

<table>
<thead>
<tr>
<th>Properties</th>
<th>BSR</th>
<th>Depolymerized BSR on different mastication time</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial plasticity (P₀)</td>
<td>26.00</td>
<td>11</td>
</tr>
<tr>
<td>Plasticity retention index (%)</td>
<td>15.40</td>
<td>18.20</td>
</tr>
<tr>
<td>Mooney viscosity</td>
<td>49.5</td>
<td>9.3</td>
</tr>
<tr>
<td>[M(1⁺+4⁺)L100°C]</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 2. The Mooney viscosity of depolymerized BSR.
The mixing time describes the time needed to homogenize the asphalt blending with asphalt modifier. The mixing time of asphalt (without modifier) is 330 min. whereas the addition of depolymerized BSR increase the mixing time of asphalt become 313-416 min. The addition of higher concentration of depolymerized BSR was also increase the mixing time of asphalt as shown in Figure 3. The increasing of mixing time was caused by the addition of asphalt modifier, which is a solid form. So that, it needs extra time to melt and blend it with asphalt.

Addition of 3% SIR 20 (without depolymerization) on asphalt needed 660 min. of mixing time (Prastanto, 2014). As compare to the addition of 3% depolymerized BSR needed 313-331 min. of mixing time. This result shows that depolymerization can reduce the mixing time of modified asphalt. The depolymerization decrease the height and weight of polymer molecules, was accordance to low P₀ and Mooney viscosity. The low P₀ was fasten the mixing time of asphalt (Cifriadi et al., 2012). The fastest mixing time is 313 minutes, which produced by 16 minutes mastication time and 3% addition of depolymerized BSR treatment. This value has decrease closely 50% of the mixing time of asphalt, which prepared by undepolymerized rubber (SIR 20).

Successful of asphalt modifier is not only showed from decreasing of mixing time but also the increasing of modified asphalt softening point. The softening point of asphalt is 49.30°C, while the softening point of modified asphalt are 49.75-54.3°C (Figure 3 & Figure 4). This result shows that depolymerized BSR can increase the softening point of asphalt, which linearly with Bakar’s et al. (2018) result. Bakar et al. (2018) reported that the addition of waste NR latex into bitumen increase the resistance of bitumen to temperature changes which refers to the increasing value of softening points. Depolymerized BSR was become matrix binder in amorphous texture of asphalt matrix. Cohesive property and plasticity of crumb rubber was delivered to crumb rubber modified asphalt (Huang et al., 2001). Those reasons related to the increasing of softening point.

### Table 3. Softening point of modified asphalt.

<table>
<thead>
<tr>
<th>Polymer</th>
<th>Treatment</th>
<th>Softening point (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BSR</td>
<td>Depolymerized on 16 minutes of mastication time. 3% depolymerized BSR was added using wet process</td>
<td>54.3</td>
</tr>
<tr>
<td>SIR 20</td>
<td>Depolymerized on 8 minutes of mastication time. 7% depolymerized SIR 20 was added using wet process</td>
<td>56.5 ¹)</td>
</tr>
<tr>
<td>SBS</td>
<td>6% SBS was added using wet process</td>
<td>68.6 ²)</td>
</tr>
<tr>
<td>CR (scrap tire)</td>
<td>Crumb rubber modified asphalt was prepare by traditional wet process</td>
<td>49.8 ³)</td>
</tr>
<tr>
<td>Concentrated latex</td>
<td>13% by total weight of Concentrated latex (60% DRC) mix into liquid asphalt</td>
<td>60 ⁴)</td>
</tr>
</tbody>
</table>

Remarks: ¹)Prastanto (2014)
²)Kök & Çolak (2011)
³)Xiang et al. (2009)
⁴)Tuntiworawit et al. (2005)

![Figure 3. Physical properties of the addition of 3% depolymerized BSR.](image-url)
The highest softening point is reached by the addition of 7% depolymerized BSR, which is masticated for 16 minutes. This value meets the minimum softening point of polymer asphalt standard (SNI 6749:2008), which is 54°C. Compared with other research that used other polymer as asphalt binder (Table 3), the softening point of depolymerized BSR is higher than scrap tire (crumb rubber) and lower than depolymerized SIR 20, concentrated latex and SBS. In general, this research describes that addition of depolymerized BSR concentration decrease the softening point as shown in Figure 2. This test result is agree well with the research test of Chen et al. (2002). The addition of SBS concentration until 9% was made a mixture of asphalt and SBS dominant. A large degree of incompatibility is shown in the large polymer domain morphologically, which causes a decrease in engineering properties due to differences in molecular weight, polarity, and structure, chemical inequalities between bitumen and SBS. Morphology is the result of the reciprocal interaction of SBS and asphalt and, consequently, is influenced by the composition of the asphalt and the nature of the polymer and content.

CONCLUSION
BSR has quietly met a standard of SIR, which appropriate of the ash and dirt content. Depolymerization is done successfully, decreases 73% P0 become 7, decreases 93.5% Mooney
viscosity become 3.2 [M (1'+4') L100°C], and increases 85% PRI become 28.6. The decreasing of P0 fastens the mixing time of depolymerized BSR and asphalt. However, addition of depolymerized BSR concentration decreases the softening point. The optimum composition of asphalt modifier is 3% addition of depolymerized BSR (16 minutes mastication time). The optimum composition reduces 347 minutes of mixing time and increases 2.2°C of softening point.

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