Modification of emulsified bitumen using Styrene-Butadiene Rubber (SBR)

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Abstract. This paper evaluates the performance of using different percentage of styrene-butadiene rubber (SBR) in rapid-setting emulsion (RS-2K). The polymer modified bitumen emulsions were blended at various percentages of SBR, i.e. 5%, 7%, 9%, 11% and 13%. The investigation focused on the optimum percentage of SBR added into the polymer modified bitumen emulsion and the performance of the polymer modified bitumen emulsion. There are several laboratory tests involved such as penetration test, softening point test and dynamic shear rheometer test that were done in order to determine the physical and rheological properties of bitumen emulsion. Based on the results, it shows that the physical properties of the polymer modified bitumen improved compared to unmodified bitumen emulsion. For rheological properties, by adding the SBR in the bitumen emulsion, the rutting resistance of the bitumen emulsion is improved. From the isochronal curve, the polymer modified bitumen emulsion shows improvement in the complex modulus, G* as comparing to unmodified bitumen emulsion. The optimum percentage of the SBR is 13% as it shows better performance compared to other percentages.

1. Introduction

Over the year, the growth of the economy of the country had caused the increment of transport on the road. With the dramatic increment of the transport on the road, the road structure has deflected and deteriorates before it achieves its design life. There are few factors that caused the road structure has deterioration which is traffic loadings and environmental factors. To minimize the deterioration, polymer modified bitumen emulsion (PMBE) is introduced in the road construction. There are numbers of application of PMBE in road construction which is used to facilitate the bonding of the old pavement and the new course which is placed over the old pavement. Besides that, PMBE can be utilized in the maintenance of the road. By using PMBE, the pavement performance can be improved and the service life is also increased as it has thermal resistance and prevents the road from rutting [1, 2]. PMBE used in a cold paving technology. It is eco-friendly as it does not require extra heat before apply it on the road surface. On the other hand, the heating process will cause environmental pollution as dust and poisonous fume will be generated during the heating process. PMBE can enhance the physical properties, durability and performance of the road.

To understand what bitumen emulsion is, it consists of a combination of water, bitumen and emulsifying agent. Bitumen emulsion contains about 50%-75% of bitumen meanwhile the water phase consists of water, acid, emulsifiers and sometimes other components. It may contain minerals which
can affect the production of bitumen emulsion. As bitumen is oil base material, water can’t mix with it easily. Hence some physical and chemical properties should be applied to them in order to form bitumen emulsion. One of the methods that allow mixing of bitumen and water is using an emulsifying agent or known as emulsifier [3, 4]. The performance of the emulsion is depends on the blending method of bitumen emulsion with polymer [5].

Bitumen emulsion is a dispersion of bitumen in an aqueous phase. Normally it is stabilized by adding an emulsifier into it. Oil-in-water (O/W) emulsion has a continuous phase in water while water-in-oil (W/O) emulsion is inverted to oil in water emulsion [6]. There are three types of setting time which are rapid setting emulsion (RS), medium setting emulsion (MS), and slow setting emulsion (SS) [7]. Each of the categories has different behavior as the setting time and percentage of bitumen content is various. In this study, rapid setting cationic bitumen is produced and characterized.

PMBE is the addition of the polymer in the bitumen emulsion in order to improve physical properties, performance and durability. Blending the bitumen with additives allows improve the range of temperature of modified bitumen and reduce temperature of the susceptibility [8]. The results of adding polymer into bitumen emulsion is that bitumen will become less brittle in low temperature and resistance to cracking. Besides, bitumen will also stiffer at high temperature to resist rutting of the pavement [9]. The polymer used to modify the bitumen emulsion is generally separated into two types which are plastomer and elastomer. Plastomer polymer can have high strength at a rapid rate but it is brittle and deform at low temperature [10]. Examples of plastomer are ethylene vinyl acetate (EVA). Unlike plastomer, elastomer can be stretched up to ten times without break and once the load is removed it will go back to original position [11]. The example of the elastomer is synthetic rubber, styrene-butadiene rubber (SBR) and crumb rubber modifier (CRM). The polymer can change the physical properties of the final product in term of the elasticity, elastomerically and viscosity [6]. Studies showed that PMBE with SBR increase ductility value [2]. Besides that, the quality of PMBE depends on the uniformity of the mixture and also type and nature of the polymer.

There are few methods to add polymer into PMBE. The method that adds polymer into the bitumen emulsion is important as it will affect the performance of the bitumen emulsion. The four methods introduce by the researcher as example pre-blending, co-milling, soap pre-batching and post blending [11]. In the post-blending method, the polymer is added into the emulsifier solution before the processing either at plant or in the field. For pre-blending method, the polymer is added into the bitumen to emulsification. The solid form of the polymer is required in this method. Co-milling is the method that separate streams of polymer, bitumen and emulsified are co-milled together simultaneously. For soap pre-batching, the polymer modifier added into soap solution which is water and emulsifier which prior to milling with the bitumen. The method to add the polymer into the bitumen emulsion is also determined by the physical form of the polymer [11].

Recent application of PMBE is widely used in road construction. There is some reason that PMBE is widely used as compared to the unmodified bitumen emulsion. PMBE can be use as preventive and corrective maintenance on the concrete pavement and asphalt pavement. Its function included stabilizing and reclaiming bases. PMBE is more economical and environment as it increases the service life of the pavement. The application of PMBE is tack coat, chip seal, slurry seal and microsurfacing. There is a lot of previous study which introduced SBR into bitumen emulsion. Therefore, this study tried to determine the optimum percentage of SBR modified bitumen emulsion and its performance by using rapid setting bitumen emulsion.

2. Materials and Methodology
This research consists of two phases. Phase I is sample preparation while phase II is material evaluation and characterization. Then followed by the grid analysis and the last stage were the conclusion and recommendation.
2.1 Sample Preparation

SBR polymer modified bitumen was prepared by using soap pre-batching method which the polymer modifier was added into the soap solution and milling with the bitumen. This emulsion was prepared by using 60/70 PEN grade bitumen. To make the emulsion, 60% of the emulsion contains of the bitumen while the remaining 40% was soap solution. Soap solutions compositions are water, emulsifier, SBR and hydrochloric acid. First of all, bitumen was heated in the oven at 150°C for melting purposes. For water phase, soap solution was prepared and temperature is maintained at 60°C. Emulsifier and hydrochloric acid were added into water and stirred. The acidity of the soap solution was checked using litmus paper and ensured to be at pH 1-2. Different percentage of SBR 1%, 3%, 5%, 7% and 11% were added into soap solution in this phase with 0% SBR as control. Emulsification process begins when both soap solution and bitumen is ready. Bitumen and soap solution were poured in the colloid mill simultaneously for 2-3 minutes for it to dispersed and gain homogeneity.

2.2 Material Evaluation and Characterization

The SBR modified bitumen emulsion underwent few laboratories testing to determine the physical and rheological properties. There were 5 tests carried out in this research which are penetration test (ASTM D5), softening point test (ASTM D36), dynamic shear rheometer test (DSR) (AASHTO T315), particle charge test (D7402-09) and residue by evaporation test (ASTM D6934-08).

2.2.1 Dynamic Shear Rheometer. The dynamic shear rheometer test is used to determine the viscosity and elastic behavior of the residue bitumen. The sample was prepared in 25 mm diameters and 1 mm thickness. The sample was tested at the oscillation speed of frequency 10 rad/s which is approximately 1.592 Hz at various temperatures. The residue of the bitumen emulsion was tested at a temperature of 46°C until the residue bitumen emulsion fails at the temperature. From the DSR the complex modulus, phase angle and the failure temperature of the residue bitumen can be determined.

2.2.2 Particle Charge for Emulsified Asphalt. This test is used to identify the charge of the emulsion. Two electrodes were inserted in the beaker which contains 250 mL bitumen emulsion. The test was started with at least 8mA of direct current connected to two electrodes. When the currents reading drops to 2mA or at the end of 30 minutes, whichever comes first, the current source disconnected and electrodes washed using distilled water. The asphalt deposited on the cathode confirmed as a cationic emulsion.

2.2.3 Residue by Evaporation. The test method is to determine the percentage of emulsion residue. 50g of bitumen emulsion sample was poured into the glass beaker with rod and weighed. The beaker was put into the oven with the temperature of 163°C ± 3C for 2 hours. After 2 hours, the emulsion was stirred and heated in the oven for another 1 hour. After 3 hours, the residue was let cool in room temperature and pour in a container for residue testing. Two testing were done on the residue of the emulsion which were penetration test and softening point test.

2.2.3.1 Penetration Test. The penetration of a bituminous material is tested for the consistency of the bitumen. The residue bitumen was placed in the water bath at 25°C for 30 minutes before the test start. The standard penetration was used; the standard needles under 100g and load for 5 seconds on the 25°C residue bitumen. Three determinations were made on the residue bitumen. For making repeated determination, the tip of the needle was pointed at for at least 10mm apart from the side of the container. The value of penetration was recorded.

2.2.3.2 Softening test. Softening test is the mean temperature at which bitumen soften and sag downwards at a distance of 25mm under the weight of a steel ball using ring ball apparatus. The residue bitumen was prepared in the dimensional brass rings and the temperature of the sample was maintained at 10°C for at least 30 minutes. The apparatus was assembled with the specimen rings, ball
centering guide and the thermometer in position and filled the bath at 105 ± 3mm with the apparatus in place. A steel ball size of 9.5 mm was placed on the centered of bitumen specimen and the test was started. The temperature was recorded when the bitumen touched the base plate.

2.2.4 Grid Analysis. Grid analysis is one of the methods that use to determine the performance of the binder. The analysis is useful when there are numbers of alternative to select with many factors to consider. This analysis was made in a table form. The type of the sample was listed as a row label and the performance tests as a column heading. In this study, each sample was assessed based on the ranking of the representative test. This method was used to determine the best performance and the optimum percentage of SBR that going to be added into the bitumen emulsion. Every sample is given a score for each test. The sample which scored the highest marks and gets the highest ranking in the table was considered the best sample. The score in the test was based on the performance of the bitumen emulsion which added different percentage of SBR and the control bitumen emulsion.

3. Results and Discussions
The results of the laboratory testing in determining the physical and rheological properties of the PMBE are discussed in detailed. The optimum percentage of modifier in PMBE is further discussed.

3.1 Particle Charge
All sample of SBR bitumen emulsion in this research have positively charged which confirmed the produced emulsions are cationic emulsion. The bitumen emulsion with the negatively charged will be compatible with the aggregate with a positive charge while the bitumen with a positive charge will be compatible with the aggregate with negatively charged.

3.2 Residue by Evaporation
Residue by evaporation test is used to determine the percentage of residue bitumen in the bitumen emulsion. The bitumen emulsion sample in this experiment is RS-2K which means it had 60% of bitumen content. The residue of bitumen emulsion will be undergoing penetration test, softening test and dynamic shear rheometer test. The specification of the bitumen emulsion has to meet the ASTM specification. From the result of testing, bitumen emulsion with 0% SBR has gotten 57% of residue bitumen, bitumen emulsion with 5% SBR has 54.84% of residue bitumen, bitumen emulsion with 7% SBR has 62.53% of residue bitumen, bitumen emulsion with 9% SBR has 59.74% of residue bitumen, bitumen emulsion with 11% SBR has 65.10% of the residue bitumen and the bitumen emulsion with 13% of SBR has 66.11% of residue bitumen.

3.3 Penetration
Penetration test is the test which used to determine the consistency of the bitumen. The sample which used on the penetration test is the sample from the residue of bitumen emulsion. Figure 1 shows the penetration value of bitumen emulsion residue with different percentage of bitumen. From the figure, 13% SBR residue bitumen emulsion has the lowest penetration value which is 40.6 PEN. The hardest bitumen is the bitumen emulsion residue with 13% SBR and the softest bitumen is the residue bitumen emulsion with 0% SBR. With the increment SBR, the behaviour of the bitumen becomes stiffer. This shows that with increasing of SBR percentage, the bitumen emulsion’s residue have turns harder that controls.
Figure 1. Penetration value of bitumen emulsion residue.

3.4 Penetration Index

The penetration index of the bitumen is used to determine the temperature sensitivity of the bitumen. The bitumen with lower penetration index will change its consistency as the temperature change. Figure 2 shows the penetration index value for bitumen emulsion residue with different percentage of SBR. Bitumen emulsion residue with 13% SBR has the lowest number of penetration index while bitumen emulsion residue with 0% SBR has the highest number of penetration index. The higher the percentage of SBR added into the PMBE, the lower the penetration index.

Figure 2. Penetration index value for bitumen emulsion residue with different percentage of SBR.

3.5 Softening Point

Softening test is used to determine the rheological properties of asphalt or residue at elevated temperature. Figure 3 shows the softening point of bitumen emulsion residue. The bitumen emulsion residue with 13% SBR has the highest average temperature which is 79.5°C while bitumen emulsion residue with 0% SBR has the lowest softening temperature which is 47.5°C. The average temperature is increasing with the increment of percentage of SBR in bitumen. From Figure 1 and Figure 3, the result of the penetration test is inversely proportional to the result of softening test. The higher the PEN
value, the lower the average temperature of the softening test which means that the bitumen sample is soft and only low temperature is required to heat and melt it.

![Softening point of bitumen emulsion residue](image1)

**Figure 3.** Softening point of bitumen emulsion residue.

### 3.6 Dynamic Shear Rheometer

DSR is used to measure the complex modulus, $G^*$ and phase angle, $\delta$ at various temperatures. $G^*$ is the sample resistance to the deformation when the sample is subjected to the repeated shear. The phase angle is the lag between the applied shear stress and resulting in shear strain.

#### 3.6.1 Isochronal Curve

The isochronal curve shows the relationship of the $G^*$ and $\delta$ at various temperatures. Figure 4 shows that the bitumen emulsion residue with 13% SBR was stiffer comparatively to other samples. At 46°C, bitumen emulsion residue with 5% SBR considered as stiffer as it has the highest value of $G^*$ among other bitumen emulsion residue. The rheological properties of the bitumen emulsion residue also determine by the phase angle. From figure 1, bitumen emulsion residue with 0% SBR shows the highest number of phase angle follows by 9% SBR, 13% SBR, 5% SBR, 7% SBR and 11% SBR.

![Relationship between $G^*$ and $\delta$](image2)

**Figure 4.** Relationship between $G^*$ and $\delta$. 
The addition of SBR can increase the stiffness of the binder residue but reduce the temperature susceptibility [2]. The addition of polymer in the bitumen emulsion can increase the rheological properties of the bitumen emulsion [12].

3.6.2 Rutting Resistance and Failure Temperature. The value of \(G^*/\sin\delta\) which determine from the DSR is used to determine the rutting resistance while \(G^*\cdot\sin\delta\) is used to determine the fatigue resistance. Bitumen is considered failed at high temperature when the value of \(G^*/\sin\delta\) below 1.0 kPa. Figure 5 shows the result of the rutting resistance of the bitumen emulsion residue. The test was started at 46°C and continue to the next temperature until the value of \(G^*/\sin\delta\) is less than 1.0 kPa which was the specification of the Strategic Highway Research program (SHRP). From the Figure 5, the bitumen emulsion residue with 5% SBR has the highest value of \(G^*/\sin\delta\) which can be explained that it is more viscous and stiffer. Among the five sample of PMBE, the bitumen emulsion residue with 0% SBR has the lowest value of \(G^*/\sin\delta\). By adding several thermoplastic or elastomer into bitumen can improve the properties of the bitumen [13].

![Graph showing the rutting criteria, \(G^*/\sin\delta\).](image)

Figure 5. Rutting criteria, \(G^*/\sin\delta\).

Table 1 shows the failure temperature of the bitumen from DSR test. The bitumen emulsion residue with 5% SBR, 11% SBR and 13% SBR has the highest failure temperature which is more than 95°C. The failure temperature does not have the exact amount as the maximum capacity of the DSR machine is 95°C. The bitumen emulsion residue with the highest failure temperature shows that the bitumen emulsion residue becomes less susceptible to permanent deformation at high temperature [14].
Table 1. Failure temperature of the binder.

| Type   | Failure temperature(°C) | G*/ sinδ = 1.0 kPa |
|--------|-------------------------|--------------------|
| 0% SBR | 74.79                   |                    |
| 5% SBR | >95                     |                    |
| 7% SBR | 87.36                   |                    |
| 9% SBR | 86.70                   |                    |
| 11% SBR| >95                     |                    |
| 13% SBR| >95                     |                    |

3.7 Grid Analysis

From Table 2, the PMBE with 13% SBR archive the highest ranking followed by 11% SBR, 9% SBR, 5% SBR, 7% SBR and 0% SBR. However, the analysis was one of the ways to determine the most suitable PMBE for tack coat, prime coat and cold mix asphalt by giving the score on the characteristic of the bitumen emulsion. From the analysis the most suitable type of emulsion is PMBE with 13% SBR which ranked first among the other sample. This is because it has the lowest penetration value and highest softening point. Besides that, it has the highest failure temperature. Temperature susceptibility of the PMBE with 13% SBR is low due to high failure temperature and ranking is based on the physical and rheological of PMBE only.

Table 2. Ranking and score of the sample.

| Type   | Physical Properties | Rheological Properties | Total score | Ranking |
|--------|---------------------|------------------------|-------------|---------|
|        | Penetration (PEN)   | Softening point (°C)   | Failure Temperature (°C) |           |
| 0% SBR | 6                   | 6                      | 6           | 18      | 6       |
| 5% SBR | 5                   | 5                      | 1           | 11      | 4       |
| 7% SBR | 4                   | 4                      | 5           | 13      | 5       |
| 9% SBR | 3                   | 3                      | 4           | 10      | 3       |
| 11% SBR| 2                   | 2                      | 1           | 5       | 2       |
| 13% SBR| 1                   | 1                      | 1           | 3       | 1       |

4. Conclusions

In this paper, the behaviour of the PMBE investigated using few laboratory tests such as penetration test, softening point test, particle charge test and dynamic shear rheometer test. Based on the result and analysis, there are few conclusions can be made:

1. The optimum percentage modifier in the SBR modified bitumen emulsion is 13%. From the laboratory testing SBR modified bitumen emulsion with 13% SBR showed better performance compares to others.

2. The modification of the bitumen emulsion with polymer improves the durability against rutting in which polymer improve the elasticity of the bitumen and finally can contribute towards the utilization of the new pavement.

3. The performance of the SBR modified bitumen emulsion improves by polymer in the bitumen emulsion. The rheological properties of the bitumen emulsion are improved in term of the complex modulus when the percentage of SBR increased. For the phase angle of bitumen emulsion is reduced with the increasing of the polymer in the bitumen emulsion.
5. References

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