

Effect of Amine Additive on Rutting Resistance of Rubberized Asphalt Binder

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ARTICLE INFO	ABSTRACT
Article history: Received 28 May 2024 Received in revised form 16 July 2024 Accepted 25 July 2024 Available online 30 August 2024 <i>Keywords:</i> Modified asphalt binder; rubberised asphalt; 3-(Triethoxysilyl)propylamine; rheology: natural rubber	Rutting is the most common asphalt pavement distress due to traffic increases. Rubberised asphalt binder was then introduced as potential materials to improve rutting resistance of the asphalt mixture. This study aims to evaluate a potential of 3- (Triethoxysilyl)propylamine as an additive for rubberised asphalt binder to improve rutting resistance. It focused on modifying the rubberised asphalt binder with approximately 0.5%, 1.0% and 1.5% of the 3-(Triethoxysilyl)propylamine. The asphalt binder was tested for penetration, softening point, rotational viscosity and dynamic shear rheometer. The results found that adding 0.5% of 3-(Triethoxysilyl)propylamine into 5% rubberised asphalt binder was significantly increased its complex modulus compare to rubberised asphalt binder without 3-(Triethoxysilyl)propylamine. It is indicating a better rutting resistance without reducing its work ability as the viscosity value is adjacent. Incorporating higher 3-(Triethoxysilyl)propylamine is not preferring as the viscosity was significantly increasing that would increase mixture production and reduced the work ability. In conclusion, asphalt binder containing 0.5% amine could be able to produce asphalt mixture with low mixing and compaction temperature and better rutting resistance. Besides, it is a potential additive for producing a sustainable asphalt binder.

1. Introduction

Rutting is a pavement distress which can occur due to traffic loading. Among pavement materials, asphalt binder at intermediate and hot temperature are mostly affecting pavement resistance to rutting. At air temperature reaches 30°C, the asphalt surface can rise to 50°C that reaches or exceeds the softening point of the asphalt binder as mentioned in [1]. As this temperature or higher is frequently experienced by most countries, improvement on the material for pavement, especially asphalt binder is important to reduce this distress to occur. Thus, a few modifiers or additives have been used lately such as plastic, natural rubber (NR), crumb rubber (CR), synthetic rubber etc. to

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improve asphalt binder high temperature properties as taken from previous study [2-5]. The NR is obtained in the form of solid particles suspended in a milky white liquid called latex, while CR is produced by removing steel and fluff by using a granular and/or cracker mill, aided by cryogenic or mechanical means of decreasing the size of the rubber particle as referred to [6]. Besides, synthetic rubber is a polymer synthesized from petroleum byproducts.

In accordance, rubberized asphalt binder is formed by adding NR, CR or synthetic rubber modifier into heated base asphalt binder. This binder then mixed with aggregate for producing the rubberised asphalt mixture. The preparation of rubberized asphalt mixture can involve two process which are wet process and dry process. Wet process is a method that blends the rubber with asphalt binder before it combines with aggregate and dry process is a method that mixed the rubber with aggregate then mixed with the asphalt binder.

There are many benefits that can be found by using rubberized asphalt. The main benefit is allowing for proper disposal and reuse of a huge number of waste rubber; and increasing demands for NR. Due to this reason, rubberized asphalt has acquired significant attention for its production and application as taken from [7]. CR has been proposed for the pavement because it is a waste material and crumb rubbers modify asphalt (CRMA) shows the greatest performances such as improving rutting resistance of asphalt mixtures when the CR percentages increase, decrease the thermal sensitivity of hot mix asphalt (HMA) and enhance resistance to permanent deformation. Besides, it reduced fatigue cracking at intermediate temperatures, thermal cracking and minimized potential of age hardening, long term maintenance costs compared to HMA as stated in [8]. Other than that, it has been found that rubberized asphalt can increase the high temperature properties; improving asphalt pavement skid resistance, road-tire noise and construction cost as mentioned in [9].

Besides, waste NR latex was reported improved the rheology properties, which indicates by improving of rutting factor (G*/sin δ) and potential to resist deformation on road surface despite of high traffic loading as claimed in [10]. Although, applying rubberized modified asphalt in a mixture needs higher temperatures for mixing and compaction compared to conventional HMA as claimed in [6,11]. These high temperatures need to ensure asphalt binder has lower viscosity and softer, high work ability, compatibility and get better coating with the aggregate. However, when the temperature is too high during mixing it will result in less fatigue resistance [12]. Other than that, higher heating processes need a great deal of energy and fuel consumption; will release toxic fumes and greenhouse gas emissions which are harmful to the environment, workers and peoples around as found by [13].

In addition, earlier in 2004, Thompson and Xiao in [14] found that a suitable blending temperature for rubberized asphalt is 177°C and reaction time of 30 minutes in the wet process. Amine-based liquid additives were used to facilitate rubberized asphalt interaction by preventing the increases in viscosity. These amine compounds facilitate the release of rubber polymer into asphalt binder, hence the overall softening of asphalt-rubber matrix. The viscosity threshold could be met at lower temperatures. This will facilitate pumping and hence allows for application of higher percentages of rubber into asphalt binder as referred to [15].

Presumably, higher application of rubber in modifying asphalt binder would be a better benefit when high proportion is used further enhance pavement performance. However, applying rubberized modified asphalt in a mixture needs higher temperatures for mixing and compaction and increase the use of energy. Therefore, this study was conducted to optimise the use of NR in rubberised asphalt binder by improving its physical and rheological characteristic to improve rutting resistance meanwhile applying amine additive in order to control the increasing of production temperature. It is expecting that high proportion of NR would be used in the mixture without significantly increasing the production temperature. The outcome of this study would be used to improve the sustainablity of the rubberised asphalt binder for a sustainable road construction.

2. Material and Its Properties

The natural rubber, 3-(Triethoxysilyl)propylamine (3-tri) and base asphalt binder penetration grade 60/70 have been used in this study. The amine was selected due to its potential to be homogenised, reduced viscosity, and enhanced adhesion of modified asphalt binders. Granite aggregate was obtained from the Hanson Quarry, Malaysia. The base asphalt binder was obtained from the Kemaman Bitumen Company (KBC), Malaysia.

3. Methodology

In this study, the rubberized asphalt was prepared by using wet mixing process. The percentage NR used is approximately 3.0%, 5.0% and 7.0% and 3-tri additives are 0.5%, 1.0% and 1.5%. All base asphalt binder and NR were mixed using high shear mixer with 1500 rpm shearing speed. The mixing procedure began with heating 420 g of base asphalt binder at temperature 145 \pm 5°C. When the temperature reached 145°C, the NR and 3-tri additives with respective percentages were incorporated into a hot asphalt binder and stirred for 45 minutes.

3.1 Physical Characteristic of Asphalt Binder Tests

The penetration test was conducted to measure the consistency or hardness of the asphalt binder under a specific test condition according to ASTM-D5 (standard test method for penetration of asphalt binder materials) specifications as referred in [16]. The higher value penetration shows that the sample is soft while the lowest value of penetration shows that the sample is hard. The softening point test was conducted to determine the temperature at which the asphalt binder became fluid and the maximum elongation distance of the asphalt binder at a specific rate and temperature. The test was conducted in accordance with ASTM-D36 (Standard Test Method for Softening Point of asphalt binder or Ring and Ball test) specifications as referred in [17] and fulfill the specifications of JKR/SPJ/2008-S4 as referred in [18].

Rotational Viscosity (RV) test was conducted to determine the viscosity of asphalt binders at high temperature range of manufacturing and construction according to ASTM D4402 as referred in [19]. The test was conducted at temperatures from 120°C to 195°C.

3.3 Dynamic Shear Rheometer (DSR) Test

Rheological properties test had been performed on rubberized asphalt binder with 3-tri additives. The complex shear modulus and phase angle were first obtained to identify the rheological properties of G*/sin δ which indicate resistance to rutting as referred to several study by [9, 11, 14, 20]. This test was carried out according to the ASTM-D7175 standard test procedures as referred in [21]. In accordance, the G*/sin δ of the rubberized asphalt binder was determined at temperature 46°C to 82°C. To resist rutting, rubberized asphalt binder should be stiff and elastic whereas the complex shear modulus elastic portion should be large [22]. The higher the G*/sin δ value, the stiffer the rubberized asphalt binder.

4. Results and Discussion

4.1 Softening Point and Penetration Value

Changes in the penetration and softening point after modification are a clear indication that the modified asphalt binders were stiffening due to the additional material. This would improve the high temperature resistance of the asphalt mixtures. Figure 1 shows the softening point of rubberized asphalt binder. The bar chart shows the rubberized asphalt binder has a higher softening point compared to base asphalt binder. The higher the percent of NR incorporated into the base asphalt binder; the higher the softening point value was measured. In accordance, the softening point of the asphalt binder incorporated 7.0% NR is the highest compared to the softening point of the asphalt binder incorporated 3.0% and 5.0% NR. Adding 7.0% NR into the base asphalt binder increased the temperature from 48.8°C to 58.8°C. That is approximately 16.9% increase and the highest among the samples that have been tested. It is clearly show that the binder incorporating 7.0% NR is harder that indicating required higher temperature to reach the softening point. Besides, Figure 2 shows the penetration value for the asphalt binder containing a different percentage of NR. The bar charts indicate the decrease of penetration value when each percent of NR is added into the base asphalt binder. In accordance, asphalt binder with 7.0% NR content has the lowest penetration value compared to asphalt binder with 3.0% and 5.0% NR content which has decreased approximately 37.5% from 69.60 dmm to 43.50 dmm. It is indicating higher NR content in asphalt binders increased the asphaltene/resins ratio and improves the stiffening properties of modified asphalt binders as also found in [23].

The penetration index (PI) was calculated to evaluate a susceptibility of the asphalt binders. The lower PI attributed to higher temperature susceptibility in asphalt binders and more cracking or rutting in asphalt mixtures. From the results, it was obtained that incorporating 5.0% NR into base asphalt binder was lowering the PI value the most, approximately 0.32 compared to other asphalt binders approximately 0.64 and 0.44 for asphalt binder comprising 3.0% and 7.0% NR respectively. It is means, asphalt binder incorporating 3.0% and 7.0% NR are lower temperature susceptibility that is attributed to less cracking and rutting potential. Besides, the effect of softening point was more pronounced for asphalt incorporating 5.0% NR that contributed to its higher PI value.



4.1.1 Rotational viscosity test

Figure 3 and Figure 4 show the viscosity of the base asphalt binder and rubberized asphalt binder; and straight line is referring the value of 3000 mPa.s according to the standard specification. It is clearly seen the same pattern for all asphalt binders which is viscosity of the asphalt binders

decreasing as the temperature increasing. The maximum asphalt binder's viscosity was observed when temperature equivalent to 120°C. For base asphalt binder, at testing temperature 120°C the viscosity is 1761.6 mPa-s and gradually decreasing approximately 87.8% toward temperature 165°C. In comparison, for NR asphalt binders the viscosity at 120°C for asphalt binder incorporating 7.0% NR was increased approximately 67.7%, followed by 5.0% and 3.0% NR which increased approximately 60.5% and 43.5% respectively higher than the base asphalt binder. Besides, at temperature 165°C, asphalt binder incorporating 3.0%, 5.0% and 7.0% NR are 48.1%, 62.1% and 76.4% respectively higher than the viscosity of the base asphalt binder. As shown in the Figure 4, all binders lower than 3000 mPa-s at 135°C that clearly fulfilled the standard specification.



Fig. 3. Viscosity of the asphalt binders against temperature



4.1.2 Effect of natural rubber content to complex modulus (G^*) and phase angle (δ)

Figure 5 shows the rheological characteristic of the base and rubberized asphalt binders. The complex modulus (G*) and phase angle (δ) of the base asphalt binder is 0.886 kPa and 86.63° respectively. As shown in the figure, at temperature 46°C to 82°C, the G* of the asphalt binders decreased which could be explained by the effect of rubber particles in the matrix as also mentioned in [9, 22]. Asphalt binder that contains 7.0% NR has the highest G* approximately 2.039 kPa and lowest δ approximately 78.36° when compared to other asphalt binders. In accordance, compared to base asphalt binder the G* of the asphalt binder with 7% NR increases approximately 56.5% while the δ decreases by 9.5%. Besides, the G* of the asphalt binder with 5% NR shows the higher value approximately 1.312 kPa and the δ is lower about 83.0° when compared with the base and 3% NR asphalt binders. The G* and δ of the asphalt binder with 5.0% NR increase by 32.5% and decrease by 4.2%. Based on the results, asphalt binder with 7.0% NR shows better rutting resistance when compared to other asphalt binders.



Fig. 5. Effect of NR on the G* of the asphalt binders

Besides, Figure 6 shows rutting characteristic of the asphalt binders. In accordance, the G*/sin δ is used to indicate the rutting resistance of pavement. The higher the value of G*/sin δ , the higher the rutting resistance. Rutting resistance of the asphalt binders at high temperature were evaluated based on the G*/sin δ that must be more than 1.0 kPa at 76°C according to the JKR specification [JKR/SPJ/2008-S4]. From the figure it is clearly seen that G*/sin δ of the overall asphalt binders decreases as the temperature increased. Asphalt binder with 3% NR shows less resistance to rutting at high temperature compared to base and other rubberised asphalt binders. However, by adding 5% and 7% NR, the G*/sin δ value at 76°C increased and exceed the minimum value of 1.0 kPa. Therefore, incorporating 5% and 7% NR into base asphalt binder was stiffen the asphalt binders that indicating improved its rutting resistivity. Other than that, the increasing G*/sin δ value shows improvement of asphalt binder's performance grade (PG). As depicted in Table 1, asphalt binder with 5% and 7% NR was improved from PG64 to PG76. Although, to prevent pavement from easily cracks while exposed to the vehicle's load and temperate, adding 5% NR is proposed to be used. It is because asphalt binder with 7% NR has remarkable high value of G*/sin δ that may result in lower fatigue resistance.



Fig. 6. Relationship of NR between G*/sinδ of the asphalt binder at different temperatures

Binder type Temperature (°C) ($G^*/sin\delta>1$ OkPa) Performance Gra					
60/70	64	1 74	PG64		
00,70	70	0.91	1 00+		
NR 3%	70	1.89	PG70		
	76	0.99			
NR 5%	70	2.52	PG76		
	76	1.32			
	82	0.72			
NR 7%	70	3.85	PG82		
	76	2.08			
	82	1.14			

Table 1

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4.2 Characteristics of Rubberized Asphalt Incorporating 3 (Triethoxysilyl)Propylamine) 4.2.1 Physical characteristic

Table 2 shows the softening point for optimum percentage of NR that has been incorporated with 3-tri to evaluate the characteristics of hot rubberized asphalt binders. The bar indicates a general trend in which every percent of amine that is added into the 5% NR asphalt binder increases the softening point. The presence of 3-tri as an additive slightly increases the temperature compared to non-additive rubberized asphalt binder. Also shown in the table, asphalt binder with 1.5% 3-tri has the highest value of softening point compared to other asphalt binders. The rubberized asphalt binders added with 0.5% and 1.0% 3-tri show the increase in softening point from 57.60 $^{\circ}\mathrm{C}$ to 60.10 $^{\circ}\mathrm{C}$ and 57.60°C to 61.50°C which increases about 4.3% and 6.8% respectively. Meanwhile, softening point of the 5% NR asphalt binder incorporating 1.5% 3-tri increases from 57.60°C to 62.60°C or increases by 8.7%. In accordance, the more the percent of 3-tri incorporating into 5% NR asphalt binder, the softening point value was increased.

In apart, as shown in the table, the penetration value of the asphalt binders indicates a reduction of penetration value when 0.5%, 1.0% and 1.5% 3-tri incorporated into 5% NR asphalt binder. The 5% NR asphalt binder containing 1.5% 3-tri has the lower penetration value compared to base asphalt binder, 5% of NR asphalt binder and other asphalt binders containing 3-tri. It is clearly seen that asphalt binder with 0.5% 3-tri has the higher penetration value compared to binders with 1.0% and 1.5% 3-tri. By incorporating 1.5% 3-tri into 5% NR asphalt binder, the penetration value was decreasing from 46.0 dmm to 23.3 dmm or 49.3%. Besides, 5% NR asphalt binder incorporated with 1.5% 3-tri has the lowest penetration value compared to non-3-tri rubberized binder and binder containing 3-tri. It is indicating that the asphalt binder is stiffer than the base and 5% NR binders without added 3-tri. Also, it was proved by the Penetration index (PI) value that by incorporating 3tri in a 5% NR binder decreased the PI value from 0.32 to -0.07. It is indicating that these asphalt binders are less susceptible to temperature and reduce potential failure due to high temperature.

Table 2

Effect of 3-0	Triethoxysi	vl)pro	pylamine on	state	condition
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Type of Binder	Properties	3-(Triethoxysilyl)propylamine (%)			
		0	0.5	1.0	1.5
5%NR asphalt binder	Softening point (°C)	56.70	60.10	61.50	62.60
+ 3-(Triethoxysilyl)	Penetration at 25°C (dmm)	46.00	30.50	29.30	23.30
propylamine	Penetration Index	0.33	0.07-	0.11	0.14-

4.2.2 Effect of 3-(Triethoxysilyl)propylamine content on rubberized binder viscosity

Figure 7 shows the result of viscosity value at different temperatures. The trend is increasing when the percentage of 3-tri incorporated into 5% NR asphalt binder is higher, while decreasing when the temperature rises. At temperature 135°C, the viscosity of the asphalt binder containing 1.5% 3tri increases by 56.1% and then followed by the asphalt binder containing 1.0% and 0.5% 3-tri which increase by 46.1% and 9.3% respectively. These values are higher than the 5% NR asphalt binder. It shows that by adding 3-tri, the viscosity of the rubberized asphalt binders higher for every percent of increment than the original 5% NR asphalt binder without added 3-tri. At testing temperature 195°C, the asphalt binder containing 0.5% and 1.0% 3-tri have about the same viscosity of 520.8 mPa-s and 566.4 mPa-s respectively. However, viscosity of the asphalt binder containing 1.5% amine was increased from 314.4 mPa-s (original 5% NR asphalt binder) to 1038 mPa-s. In conclusion, by adding amine the viscosity of the rubberized asphalt binder is higher.

In addition, Figure 8 shows the result of viscosity at 135°C for the binder containing 0.5%, 1.0% and 1.5% 3-tri; and straight line is referring value of 3000 mPa.s according to standard specification. Based on the graph, the highest viscosity value was obtained for binder incorporating 1.5% 3-tri which is 4326 mPa-s. By adding 1.5% 3-tri into 5% NR asphalt binder, the viscosity was increased by 56.1% followed by the asphalt binder incorporating 1.0% and 0.5% 3-tri which were increased by 46.1% and 9.3% respectively. In accordance, using low 3-tri resulted in lower asphalt binder activation energy compared to using higher 3-tri [22]. Based on the specification (JKR/SPJ/2008-S4), the requirement value is lower than 3000 mPa-s at temperature 135°C. As shown in the figure, only binder incorporating 0.5% 3-tri does not exceed the maximum value. Therefore, the optimum 3-tri incorporating into the 5% NR asphalt is 0.5% to ensure the viscosity does not exceed the requirement.



temperature

Fig. 8. Viscosity of the asphalt binders at 135°C

4.2.3 Effect of 3-(Triethoxysilyl)Propylamine content to G^* and δ

From Figure 9, the result at temperature 46°C to 82°C shows that, the G* was increased while the δ was decreasing. It meant that when the temperature became higher and higher, the viscous component decreased, and the asphalt tended to become less viscous liquid. By adding 0.5% amine into the 5% NR asphalt binder, the failure temperature increases from 76°C to 82°C compared to the base 5% NR asphalt binder. Furthermore, by adding 1.5% 3-tri, the G* is 9.58 kPa and the lowest δ is 70.45° compared to other samples. Lower δ when more 3-tri was added indicated that less viscous and more elastic components existed in the binder. It also implied that binder with higher 3tri might be easier to recover after loading compared with the binder with lower 3-tri had better rutting resistance. These findings were also obtained by the previous study by Gong *et. al* in [23]. In accordance, the binder incorporating 1.5% 3-tri increases by 86.3% of G* while for δ , it decreases by 15%. For the binder incorporating 1.0% 3-tri, the higher G* is 4830 Pa and lower δ is 73.01°, while for binder incorporating 1.0% 3-tri, the G* was increased 72.9% and δ was decreased 12% compared to base 5% NR asphalt binder. Based on the result, asphalt binder incorporating 1.5% 3-tri content shows better rutting resistance when compared to the base 5% NR asphalt binder. However, asphalt binder with 0.5% 3-tri is more applicable by considering its viscosity value not exceeding 3000 mPa-s as according to the specification compare to other asphalt binders (JKR/SPJ/2008-S4).



Fig. 9. Effect of 3-(Triethoxysilyl)propylamine on G* and for asphalt binder

4.2.4 Effect of 3-(Triethoxysilyl)propylamine on rutting parameter

The higher rutting factor of the asphalt binder implied a higher percentage of elastic components existed in asphalt binder with better deformation resistance. Based on Figure 10, it is clearly shows that for all samples rutting resistance was reducing significantly when the temperature increasing. It could also be found that asphalt binder with amine had a larger rutting factor than that of the asphalt binder without amine as also revealed in [23]. That is to say, different composite additives have different degrees of improvement. Besides at each temperature, the base 5% NR asphalt binder has less capability to resist rutting compared to other 5% NR asphalt binders incorporating 3-tri. Besides, by incorporating 0.5%, 1.0% and 1.5% 3-tri in 5% NR asphalt binder, the G*/sin δ value at 76°C increase and exceed the specification which is more than 1.0 kPa. Besides it clearly shows that the G*/sin δ values for asphalt binders containing amine are still higher than 1.0 kPa at 82°C. In accordance, roughly the grade of these asphalt binders was increased from PG76 to PG82 indicating that the asphalt binders is better in rutting resistance. However, for producing asphalt mixture a low viscosity asphalt binder needs to ensure a better coating with aggregate. By comparing all binder incorporating amine, incorporating 0.5% 3-tri into base 5% NR asphalt binder is optimum dosage as the viscosity of this asphalt binder is less than the specification of 3000 mPa-s and close to the base 5% NR asphalt binder. Also, it could be highlighted that the difference between the viscosity is 10.30%. Besides, difference on G*/sin δ between base 5% NR asphalt binder and 5% NR asphalt binder with 0.5% 3-tri is significant that are 1.76 and 4.60 kPa respectively. It is indicating the 5% NR

asphalt binder incorporating 0.5% 3-tri is 248.1% better in rutting resistance. Besides, the viscosity is only different 10.30% from the viscosity of the base 5% NR asphalt binder.



different temperature for asphalt binder

5. Conclusions

It is indicating, adding 5% NR into base asphalt binder is an optimum percent to increase grade of base binder to PG76. Its rutting factor is G*/sin\delta exceeds 1.0 kPa and the viscosity is 1897.2 mPas at 135°C that is less than 3000 mPa-s. Besides, incorporating 7.0% NR reduced the work ability which is difficult to homogeneous due to foaming during the blending process and needing the highest temperature to melt the asphalt binder. For the asphalt binder incorporating 3-tri, the physical test of softening point penetration test and rotational viscosity resulted in the presence of 1.5% 3-tri as an additive has increased the softening point and reduces to the lowest the penetration value compared to other percentage of additive. In term of rheology characteristic, rubberised asphalt binder incorporating 0.5%, 1.0% and 1.5% 3-tri have increased and exceeded the specification of the G*/sin\delta value must be more than 1.0 kPa. It could be concluded that 0.5% 3-tri is the optimum content of the additive for producing rubberised asphalt binder with better rutting resistance. In accordance, adding 0.5% of 3-tri into 5% NR modified asphalt binder was significantly increased the complex modulus of the rubberised asphalt binder in therefore improve the rutting resistance. Also, its viscosity is 1897.2 mPa-s at 135°C that closed with the bane 5% NR asphalt binder. Incorporating higher 3-tri was increase the rutting resistance. Although, incorporating 3-tri higher than 0.5% into 5% NR asphalt binder is not suggested by considering the viscosity of the asphalt binder is significantly increasing that would increasing the temperature for mixture production and reduced the work ability.

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