



Article Masterbatch Natural Rubber—Innovative Asphalt Cement Additive for Sustainable Flexural Pavements

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Abstract: The mechanical performance of Masterbatch Natural Rubber (MNR)-modified asphalt concrete (MNR-AC) was investigated and is presented in this paper. When compared to conventional asphalt concrete (AC), MNR-AC exhibits significantly superior performance across key mechanical parameters, including Marshall stability, indirect tensile strength (ITS), resilient modulus (IT Mr), indirect tensile fatigue life (ITFL), and rutting resistance. The most pronounced enhancements are observed at the optimal dry rubber to asphalt cement (r/b) ratio of 3%, at which MNR-AC demonstrates peak performance in all evaluated tests. The fatigue distress models for MNR-AC and AC reveal distinct logarithmic relationships, with an intersection point occurring at an r/b ratio of approximately 3%. This suggests that MNR-AC with an r/b ratio of 3% or less exhibits a markedly superior fatigue life compared to conventional AC under equivalent applied-stress conditions. MNR offers significant practical advantages over liquid natural rubber, including more consistent mixing, and simplified storage and transportation, positioning it as a promising and sustainable advancement in pavement material technology.

Keywords: masterbatch natural rubber; asphalt concrete; fatigue model; rutting failure; resilient properties

1. Introduction

Asphalt concrete serves as a fundamental component of flexible pavement. Traditional flexible pavement offers the advantage of adaptability to underlying soil movements, effectively distributing loads across layers, and providing a smooth, durable surface suitable for various traffic conditions. The rapid advancement in global economic development has led to a substantial increase in transportation demands, particularly within the commercial sector. The escalation in the weight and volume of transported freight forwarding has significantly strained road systems, resulting in a marked rise in premature pavement distress. This premature failure not only diminishes the expected lifespan of the pavement but also escalates maintenance costs and disrupts the efficiency of transportation networks. In response to the problems presented by the increased transportation loads, several pavement engineers and researchers have sought to improve asphalt cement binders to improve the performance and durability of flexible pavements. One such advancement is



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Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). the development of polymer-modified asphalt (PMA), which has demonstrated efficacy in meeting the heightened demands of modern transportation networks [1,2].

PMA is a special type of asphalt cement binder, which incorporates synthetic polymer additives for superior property. PMA has been utilized worldwide for heavy traffic pavement surfaces, with high short-term and long-term performance. The principal mechanism driving high performances are attributed to the strengthened adhesive strength at the point of contact between the binder and aggregate, brought about by the incorporation of polymers. The integration of PMA in asphalt concrete pavement has yielded superior performance in terms of stability, durability, compressive and tensile strengths, and resistance to both rutting and fatigue cracking, when compared to conventional asphalt concrete [2–4].

Common synthetic polymer additives for modifying asphalt cement to produce PMA include thermoplastic elastomers (TE), Ethylene Vinyl Acetate (EVA), polyethylene (PE), and styrene-butadiene rubber (SBR). TE is a synthetic polymer with excellent thermoplastic and elastomeric properties, attributed to the cross-linking of compounds within a threedimensional network. Styrene–butadiene–styrene (SBS) is categorized as a TE, with a high potential to enhance asphalt cement's rheological properties. PMA with SBS also exhibits enhanced flexibility and resistance to thermal cracking, hence is preferential in regions with extreme temperature variations. In contrast, in hot climates with high traffic loads, EVA and PE, which improve thermal stability and resistance to rutting, are more suitable. SBR is classified as an elastomeric polymer that enhances the ductility of asphalt concrete pavement, thereby increasing its flexibility and resistance to cracking at low temperatures. In addition to the aforementioned additives, other types of additives such as styrene-isoprene-styrene (SIS), styrene-ethylene-butadiene-styrene (SEBS), ethylenepropylene-diene terpolymer (EPDM), isobutene-isoprene copolymer (IIP), crumb rubber, polybutadiene (PBD), and polyisoprene can also be applied as the asphalt cement modifier effectively [5–11].

Natural rubber, NR, derived from the Hevea brasiliensis tree, is a plant-based elastomer with significant versatility and value in the industrial sector. NR predominantly consists of long-chain polymer molecules, with its main structural component being polycis-1,4 isoprene, where a biopolymer is characterized by repeating isoprene monomers. Numerous studies have demonstrated the efficacy of NR as an additive for asphalt cement, identifying NR as a crucial factor in enhancing the performance of asphalt cement, including for penetration, softening point, ductility, viscosity, elastic recovery, dynamic shear modulus, and rheological properties. The enhanced asphalt cement performance is attributed to the formation of cross-linking and bridging interactions between NR and asphalt cement molecules. These interactions strengthen the mechanical bonds within the asphalt cement matrix, resulting in enhanced elastic properties compared to conventional asphalt cement [12–14].

As the world top three NR-producing countries (Thailand, Indonesia, and Malaysia), various rubberized asphalt cement technologies have been developed to address specific demands, such as enhancing the performance of asphalt cement and promoting the policy of environmental sustainability and economic benefits. Various NR additives, such as concentrated NR, pre-vulcanized liquid NR, and vulcanized liquid NR, have demonstrated significant potential for enhancing the performance of flexural pavements [15–22], similar to synthetic polymer. Udomchai et al. [23] have reported that NR promoted sustainability through the reduced thickness of asphalt concrete pavements for traffic load design, as a result of its highly resilient properties, hence reducing total construction costs compared to conventional asphalt concrete pavement.

Concentrated NR is derived from raw NR in liquid form, with a dry rubber content exceeding 60% of the total NR weight. It is stabilized with surfactants and additional additives to prevent latex coagulation and improve mechanical bonding. Pre-vulcanized NR is an enhanced form of concentrated NR, treated with vulcanizing agents to create cross-linked and bridged rubber molecules, resulting in superior adhesive properties. Vulcanized NR, the most advanced form, is produced by heating pre-vulcanized NR during the

vulcanization process, which creates exceptionally strong bonds between rubber molecules, enhancing its overall durability and stability. In terms of the application, concentrated NR offers cost-effective mechanical improvements, while pre-vulcanized NR provides better performance at a higher cost [16,17].

Most applications for the modified performance of flexible pavements worldwide have similarly relied on liquid NR. Building on the authors' successful research on liquid NR forms (concentrated NR and pre-vulcanized NR) for enhancing asphalt concrete performance, supported by the Rubber Authority of Thailand [16,17], this study carries the next step by exploring the solid form of NR. The application of solid NR on asphalt concrete mixing represents a groundbreaking advancement in Thailand that offers strategic advantages over liquid NR, particularly in terms of simplified transportation and storage.

In terms of storage, solid NR can be readily stored in conventional warehouse facilities, whereas liquid NR necessitates specialized tanks to prevent coagulation and preserve its quality. Furthermore, solid NR can be easier and safer for transportation, eliminating the need for specialized containers. This advantage significantly streamlines logistics and reduces the complexities associated with liquid NR transport.

Masterbatch Natural Rubber (MNR) is an advanced product derived from NR, presented as a concentrated mixture in solid form. MNR is produced by blending concentrated NR with specific additives and reinforcing fillers, typically carbon black, in a controlled environment to ensure uniform dispersion until coagulation is achieved. The coagulated mixture is then filtered, and dried to create a concentrated form of rubber that can be easily incorporated into various manufacturing processes. The MNR exhibits excellent mechanical properties, including high tensile strength and elasticity, as well as exceptional resistance to wear, heat, and chemicals. In Thailand, MNR is a highly valued material in the manufacturing industry, enhancing the performance and durability of products such as tires, conveyor belts, seals, gaskets, footwear, and adhesive products [12,24,25].

In-depth investigations into its potential application in asphalt concrete pavements are the focus of this research. MNR-modified asphalt was innovatively developed as a mixture of ordinary asphalt cement with a penetration grade of 60/70 and MNR. The influences of MNR content on the performance of asphalt concrete were examined in this research, comparing them with those of conventional asphalt concrete using the parameter of the dry rubber to asphalt cement (r/b) ratio. The tensile and compressive mechanical performances were examined under static and cyclic loads to simulate the actual traffic loading-induced damage to asphalt concrete pavement. The fatigue model based on a mechanistic–empirical approach was developed for MNR–asphalt concrete and compared with that for conventional asphalt concrete. The outcomes of this research will significantly advance the sustainable application of NR products in the pavement construction sector, not only in Thailand but also in other NR-producing countries worldwide.

2. Materials

The commercial Masterbatch Natural Rubber (MNR) used in this study was supplied by the Rubber Authority of Thailand and manufactured from domestic natural rubber (NR), adhering to the Thailand Industrial Standard TIS 2731/2559 [26]. The raw NR was cleaned of impurities like dirt and bark, then concentrated to achieve a dry rubber content (DRC) exceeding 60% by centrifugation, separating heavier rubber particles from lighter water and non-rubber substances. The concentrated NR was treated with surfactants, including sodium dodecyl sulfate (SDS), antioxidants, and zinc oxide, to prevent coagulation and to stabilize the rubber molecules. Reinforcing fillers, typically carbon black, were dispersed in a surfactant solution to ensure uniform distribution and prevent agglomeration. The stabilized NR was blended with the fillers at 70 °C for 1 h at 1000 rpm. The mixture underwent vulcanization with sulfur, tetramethylthiuram disulfide (TMTD), and zinc diethyldithiocarbamate (ZDEC) at 140 °C for 45 min at 1500 rpm, followed by heating at 160 °C for 1 h at 1500 rpm to further cross-link the rubber molecules. The mixture was coagulated in 90% aqueous acetic acid at room temperature with gentle stirring (300 rpm) for 1 h, and thereafter filtered and dried at 70 $^{\circ}$ C in a vacuum oven for 24 h to obtain the solid MNR.

Limestone (L), Thailand's primary aggregate and one commonly employed in the construction of flexible pavements, was used as an aggregate in this research. The L aggregate was procured from a stone mill located in Saraburi Province, Thailand. The studied aggregate was examined for its engineering properties based on the specification of the Thailand Department of Highways [27], as detailed in Table 1. The L aggregate fulfilled the requirement for flexible pavement-material selection for wearing surface courses [27].

Table 1. Properties of studied aggregate [17].

Property Measure	DH-S 408/1989 [27] Specifications	Values [17]		
Soundness (%)	<9	2.4		
Los Angeles abrasion value, LA (%)	<35	24.6		
Flakiness index (%)	<35	16.0		
Elongation index (%)	<35	20.0		
Aggregate impact value, AIV (%)	<25	20.2		
Aggregate crushing value, ACV (%)	<25	20.8		
Coating and stripping (%)	>95	98		
Asphalt absorption (%)	-	0.22		

The ordinary asphalt cement penetration grade 60/70, the primary binder for the Thailand flexible pavement construction, was selected as a control binder. The studied asphalt cement was a commercial product. Its engineering properties were found to meet the minimum standards [28], as detailed in Table 2.

Table 2. Properties of studied MNR-asphalt cement.

Properties	Unit DH	DH-SP 401/1988 [28]	Properties of Asphalt Cement [17]	Properties of MNR–Asphalt Cement at Different r/b Ratios							
		Specifications		3	5	7	8	9	11	13	15
Penetration Softening point (°C)	°C	50–70 >50	67 47.8	57 58	55 61	53 62	50 64	51 60	54 56	58 50	66 42
Flash point (°C)	°C	>220	332	250	249	247	245	245	236	231	228
Elastic recovery (%)	%	>40	35	50	46	41	37	34	29	25	21
Brookfield viscosity	mPa*s	200–600	350	354	358	361	367	387	475	511	612
Dynamic shear resistance G*/sinδ	kPa	>1.0	1.2	3.34	4.05	4.66	5.19	5.94	7.21	7.41	7.83

3. Experimental Program

3.1. Sample Preparation and Basic Properties Test

The particle-size distribution of the studied aggregate was initially assessed using the sieving method. Subsequently, its gradation curve was modified to comply with the specifications of the Marshall method based on DH-S408/1989 [27]. Figure 1 illustrates the modified gradation curve for the studied aggregate, where the solid line represents the criteria of the upper and lower limits specified by DH-S408/1989 [27]. The MNR-modified asphalt concrete (MNR-AC) and conventional asphalt concrete (AC) samples were prepared under the specifications of DH-T 604/1974 [29]. Previous studies by Hoy et al. (2024) [16] and Aiamsri et al. (2024) [17] demonstrated that incorporating a small amount of NR additive results in the best performance of NR-modified asphalt concrete. As such, the impact of MNR on the asphalt cement properties affecting the performance of asphalt concrete was systematically evaluated in small increments of the r/b ratio = 0, 3, 5, 7, 8, 9, 11, 13, and 15%. The MNR was blended with asphalt cement using a heat-stirring mix process. The mixing was performed by stirring at a frequency of 180 rpm at 170 °C, in accordance with DH-S408/1988 [28]. Subsequently, the MNR-modified asphalt cement was



1

Aggregates Particle Size (mm)

stored in a metallic container, maintained at a temperature of 170 °C, in preparation for the asphalt concrete mixing process.

Figure 1. Gradation curve of studied aggregate [16] compared to the graduation specification [27].

0.1

0.01

The L aggregate, with its modified gradation, was heated at a temperature of 160 °C. The heated aggregate was then mixed with the heated asphalt cement and MNR–asphalt cement at 150° C for 60 s. The conventional asphalt cement and MNR–asphalt cement contents were varied at levels of 4.0, 4.5, 5.0, 5.5, and 6.0% by the weight of total mixtures to find out the optimum asphalt cement content at a specific air void of 4%. The mixtures were then subsequently compacted in standard metallic molds, each with a diameter of 101.60 mm, using 75 blows on each side, in accordance with the Marshall method. At least 3 samples were prepared for each experimental test condition. The conventional asphalt cement and MNR-modified asphalt cement samples were initially assessed for engineering properties following the Marshall method. The samples were placed in a conventional Marshall stability load frame and subjected to a vertical load at a settlement rate of 50 mm/min. The vertical settlement was quantified utilizing a linear variable differential transformer (LVDT) equipped with an automated data recorder [29].

3.2. Performance Tests

100

80

60

40

20

0

100

10

Percentage of Passing (%)

The conventional AC and MNR-AC samples underwent tensile and compressive performance evaluations, encompassing both static and cyclic loadings, to derive comprehensive design parameters. The static tensile characteristic was appraised using the indirect tensile strength (ITS) test under ASTM D6931-17 [30]. Cyclic tensile performance was assessed using the tensile resilient modulus (IT Mr) and indirect tensile fatigue (ITF) tests, in accordance with the ASTM D4123-95 [31] and EN12697-24 [32] standards, respectively.

The ITS evaluation process involved applying vertical static loadings to both the AC and MNR-AC samples at a deformation rate of 50 mm/min until the samples completely failed, based on ASTM D6931-17 [30]. Prior to the test, AC and MNR-AC samples were positioned in an indirect tensile loading equipped with a 19 mm wide and 125 mm long loading strip, within a temperature-controlled chamber. The test temperature was consistently maintained at 25 °C throughout the entire duration of the test.

The determination of IT Mr involved subjecting AC and MNR-AC samples to a haversine loading pulse at a stress level of 15% of the ITS. The loading frequency was set to 1.0 Hz, with a 0.1 s loading interval and a 0.9 s rest interval, repeated for 200 cycles, following ASTM D6931-17 [30]. Horizontal deformation during each loading cycle was detected automatically using a linear variable differential transformer (LVDT).

To investigate the fatigue behavior under repeated traffic loading, the ITF testing was carried out at the imposed stress levels of 250, 300, and 350 kPa, recommended by

EN12697-24 [32]. The various repetitive stress levels were applied to the sample through the haversine load pulses at a frequency of 1.0 Hz (0.1 s loading period and 0.9 s rest period). The test continued until total failure or when vertical deformation exceeded 12 mm.

The rutting resistance of AC and MNR-AC samples was evaluated using a Hamburg wheel tracker, a widely recognized means for evaluating the moisture susceptibility and rutting resistance of asphalt concrete samples. According to AASHTO T324 [33], the AC and MNR-AC samples were immersed in water at a consistent temperature of 50 °C throughout the testing period. The samples were then subjected to a wheel load of 1.5 kN through a steel wheel (203 mm diameter, 47 mm width) at 50 °C. The rut depth was automatically measured for each wheel cycle and continuously monitored during wheel movement. The testing was automatically completed after either 10,000 cycles or upon reaching a rut depth of 20 mm. The resistance to rutting failure was evaluated based on the cumulative rut depth at the end of the testing cycle.

3.3. Fatigue Distress Model

The ITFL results were used to develop the fatigue distress model for MNR-AC, which is a fundamental tool for mechanistic pavement design. The fatigue distress model, which is the ITFL and logarithm of initial tensile strain, εt , was prescribed in EN 12697-24 [32] in the following form:

$$\text{ITFL} = a \left[\frac{1}{\varepsilon_t} \right]^b \tag{1}$$

where *a* and *b* are fatigue parameters that vary according to the material properties.

The εt can be calculated in terms of IT Mr under a specific stress level:

$$\varepsilon_t = \frac{\sigma(1+3v)}{ITM_r} \tag{2}$$

where σ is the applied stress (kPa), and v is Poisson's ratio (assumed to be 0.35 based on EN12697-24 [32]).

4. Experimental Results and Discussion

The conventional AC data from the previous study [17] were taken for comparison purposes because the data were generated under the same controlled and standardized testing conditions as this study for MNR-modified AC. This ensures consistency in the methodology, materials, and environmental factors. The impacts of the integration of MNR on the characteristics of traditional asphalt cement at various r/b ratios are summarized in Table 2.

The investigations were conducted in accordance with the standard test method for asphalt cement specification by the Department of Highways of Thailand [28]. The traditional asphalt cement meets all the minimum requirements of asphalt cement specifications [29]. Introducing MNR with an incremental r/b ratio significantly enhances the performance of asphalt cement compared to conventional asphalt cement. The incorporation of MNR contributes significantly to the formation of a polymeric network through the cross-linking of NR molecules within the asphalt cement matrix, thereby enhancing the overall elastic performance properties. The penetration values of MNR–asphalt cement consistently decrease as the r/b ratio increases, ultimately reaching the lowest value when r/b = 8%. The integration of MNR into the asphalt cement yields the highest improvement in elastic recovery at an r/b ratio of 3%. In addition, the dynamic shear resistance of asphalt cement is enhanced, with the highest value observed at r/b = 15%.

The experimental test data of the conventional AC sample, based on the previous research [16,17], were taken and reanalyzed for comparison purposes. Figure 2 depicts the correlation of Marshall stability versus the r/b ratio of MNR-AC samples. The solid line represents the minimum Marshall stability requirement (8.0 kN) specified by DH-S 408/1989 [27]. The addition of MNR improves the properties of asphalt cement, leading

to an increase in Marshall stability. The highest Marshall stability value is reached at the optimum r/b ratio of 3%. At the optimal r/b ratio, the Marshall stability value of the MNR-AC sample is 12.5 kN. Subsequently, the Marshall stability decreases as the r/b ratio exceeds the optimal value.



Figure 2. Marshall stability of MNR-AC samples at different r/b ratios [17,27].

The correlation between indirect tensile strength (ITS) and the r/b ratio of the AC and MNR-AC samples is illustrated in Figure 3. Similar to the Marshall stability results, the highest ITS values are achieved at an r/b ratio of 3%. At the optimum r/b ratio, the ITS value of the MNR-AC sample is 437.1 kPa.



Figure 3. ITS of MNR-AC samples at different r/b ratios [17].

The IT Mr results of the AC and MNR-AC samples are presented in Figure 4. Introducing the MNR contributes to enhanced resistance to plastic deformation, thereby enhancing the IT Mr. The highest IT Mr is observed at the same optimum r/b ratio of 3% (IT Mr decreases when r/b > 3%), consistent with the ITS results. This correlation indicates that the improved bonding and cohesion imparted by the r/b ratio not only strengthens the tensile strength but also enhances the asphalt concrete's overall elasticity and resilience, leading to better performance under repeated loading conditions. At the optimum r/b ratio of 3%, the IT Mr of the MNR-AC is 3896 MPa, which is a 26% improvement compared to the conventional AC sample.



Figure 4. Relationship between IT Mr and r/b ratio of MNR-AC samples [17].

Figure 5a–c presents the evaluated results of the indirect tensile fatigue life (ITFL) of AC and MNR-AC samples at various r/b ratios under the imposed stress levels of 250, 300, and 350 kPa. The higher applied stress causes a lower ITFL in all experimental samples due to a higher rate of plastic deformation development.



Figure 5. Cont.



Figure 5. Relationship between ITFL and r/b ratio of MNR-AC samples at different r/b ratios under stress levels of (**a**) 250, (**b**) 300, and (**c**) 350 kPa [17].

At a specific stress level, the enhanced resilient performance of the MNR-AC samples, characterized by a more predominant elastic response, contributes to the improved cracking resistance, thereby increasing ITFL. The highest ITFL values are observed at an r/b ratio of 3%, consistent with the results from ITS and IT Mr testing, regardless of stress level. Similar to the ITS and IT Mr test results, ITFL value decreases with an increase in the r/b ratio exceeding 3%. The improvement in ITFL can be attributed to the increased ITS and IT Mr values when MNR is incorporated. An enhanced ITS indicates better resistance to tensile stress and cracking, while improved IT Mr reflects superior elastic recovery and resilience.

A wheel tracker test was conducted to measure the irreversible deformation in asphalt concretes caused by moving wheel loads. This test simulates the accumulation of plastic strain and binder stripping, which eventually results in surface rutting [8]. Figure 6a illustrates the schematic plot depicting the correlation between rut depth and the number of wheel cycles, which is characterized by two distinct zones based on the slopes. The first zone, the creep slope, shows a linear increase in rut depth with the number of wheel cycles, attributed to the creep behavior of asphalt concretes under repeated stress.

The second zone, the stripping slope, reflects the extent of damage to the asphalt concretes, influenced by the combined impacts of moisture and temperature. The intersection between the creep slope and the stripping slope is labeled as the inflection point, marking the transition from the final stage of creeping to the initial stage of stripping. Theoretically, materials exhibiting a lower creep slope demonstrate a greater rut resistance [1]. Consequently, the relationship between the creep slope and the r/b ratio was developed and is presented in Figure 6b. The creep slope initially decreases with an increasing r/b ratio, reaching the minimum value at the optimum r/b ratio of 3%. The lowest creep slope at a 3% r/b ratio indicates the highest material stability and resistance to plastic deformation under continuous loading. At this optimum r/b, the MNR-AC is more effective at distributing and absorbing stress, thereby delaying significant rutting. As the r/b ratio surpasses 3%, the creep slope increases significantly. This increase indicates that the asphalt concrete samples are experiencing more pronounced deformation under repeated loading, meaning they become less effective at resisting the long-term accumulation of strain.

The relationship between rut depth at the end of the test and the r/b ratio of conventional AC and MNR-AC samples is shown in Figure 6c. The MNR-AC at the optimum r/b ratio demonstrates superior rutting resistance compared to the conventional AC. The minimum rut depth is observed at the optimal r/b ratio of 3%, which exhibits the lowest creep slope. When r/b > 7%, the number of wheel cycles at a 20 mm rut depth, N_w, reduces with the increased r/b ratio, indicating the reduced rutting resistance.



Figure 6. Relationship between (**a**) rut depth and number of wheel cycles in schematic plot, (**b**) creep slope and r/b ratios, and (**c**) rut depth and r/b ratio of MNR-AC samples.

The experimental findings show that the performance of the MNR-AC samples is predominantly influenced by the r/b ratio, which was found to be depended on elastic recovery. The ideal r/b ratio of 3% for the best performance is found to be closely aligned with the optimal r/b ratio at the maximum elastic recovery of 50%. Figure 7 shows the

correlations of elastic recovery versus ITS and the IT Mr of the MNR-AC samples, which can be represented by a linear function. This result confirms the direct influence of the improved elastic recovery on the improvement of the MNR-AC performances. Improved elastic recovery enhances the adhesive and cohesive properties of modified asphalt concrete, hence mechanical properties' improvement across all experimental static and cyclic tests. As such, the elastic recovery can be practically used as a prime factor to quickly determine the optimum r/b ratio of the MNR-modified asphalt cement, prior to complicated mechanical performance testing.



Figure 7. Influence of improved elastic recovery of MNR–asphalt cement on improvement of mechanical properties, such as ITS and IT Mr, of MNR-AC for all r/b ratios.

5. Distress Model for MNR-AC

The interrelationship between the IT Mr and ITS of the MNR-AC at various r/b ratios was established and was benchmarked with the test data of conventional AC (r/b = 0%), as demonstrated in Figure 8. The IT Mr exhibited a linear relationship with ITS, consistent with previous research findings on pre-vulcanized rubber latex-modified asphalt concrete [17]. The linear relationship between IT Mr and ITS, with a very high coefficient of determination ($R^2 = 0.93$), can be expressed as the following equation:

$$IT Mr = 14582(ITS) - 2703.2$$
(3)

In addition to the very high R², the linear relationship provides a reliable and straightforward predictive model for predicting the material's property under varying r/b ratios. The demonstrated correlation clearly indicates that the higher ITS is directly associated with a higher IT Mr. Thus, it can be inferred that the improved bonding strength facilitated by MNR integration enhances both ITS and IT Mr.

Using Equation (2), the strain response of MNR-AC at various r/b ratios could be calculated and shown in Figure 9. Figure 9a depicts the relationship between ε_t and the r/b ratio. It is evident that as the r/b ratio increases beyond 3%, the ε_t begins to rise sharply, particularly at the higher stress levels of 300 kPa and 350 kPa, which suggests that increasing the r/b ratio beyond this point leads to diminished performance in terms of strain resistance. This figure demonstrates that at an r/b = 3%, the initial strain remains relatively low, indicating better resistance to deformation. Figure 9b shows the relationship between ε_t and IT Mr across the same three stress levels. Notably, the sample with a higher IT Mr value exhibits significantly lower initial strain for the same stress level, which aligns with improved fatigue resistance.



Figure 8. Relationship between IT Mr and ITS of MNR-AC.



Figure 9. Relationship between (**a**) initial strain (ε_t) and r/b ratios, and (**b**) initial strain (ε_t) and IT Mr of MNR-AC samples.

The fatigue distress model for MNR-AC was established using the results from Figures 5 and 9 and is demonstrated in Figure 10. It is crucial to note that the proposed fatigue distress model for MNR-AC follows a unique logarithmic relationship for all r/b ratios tested, which is in line with the fatigue law suggested in EN 12697-24 [32]. The samples with superior IT Mr values exhibit a lower initial strain under the same applied stress, resulting in a higher ITFL.



Figure 10. Fatigue distress model of MNR-AC samples at different r/b ratios [17,34].

In Figure 10, the distress model of conventional AC (data from Aiamsri et al., 2024 [17]), is developed and plotted for comparison with that of MNR-AC. It is evident that the proposed model for conventional AC demonstrates a similar slope compared to the distress model for standard asphalt concrete described by AI, 1981 [34], but with different positions. This difference might be due to variations in material properties.

When comparing the destress model of MNR-AC with that of conventional AC, there is an intersection between these two models. The intersection point is at an r/b between 3 and 5%. This means that for the same applied stress, the MNR-AC with an r/b ratio \leq 3% has a higher ITFL than the conventional AC. In other words, the usage of an r/b > 3% is not effective when the applied stress is less than 350 kPa at a 1 Hz frequency in terms of both engineering and economic viewpoints. At these r/b ratios, the IT Mr of MNR-AC is lower than the conventional AC at the same applied stress (Figure 4). Hence, the ε_t is larger and ITFL is smaller (Figure 5).

Based on these research findings, a mechanistic pavement design using MNR-AC can be carried out using the proposed distress model, where the ε_t can be determined from IT Mr. The IT Mr can be practically approximated using the proposed empirical equation, Equation (1), if the ITS value is known.

6. Conclusions

This study evaluates the influence of Masterbatch Natural Rubber (MNR) on the performance of asphalt cement and asphalt concrete, with a particular focus on the impact of the r/b ratio. The integration of MNR into conventional asphalt cement (AC60/70) at an optimum content level resulted in measurable improvements across several performance indicators, providing a more quantifiable demonstration of its effects.

One of the key improvements observed was in Marshall stability, where MNR-modified asphalt concrete (MNR-AC) achieved a stability value of 12.5 kN at the optimum r/b ratio of 3%. This suggests stronger bonding between the asphalt cement and aggregates due to the inclusion of MNR.

At the optimal r/b ratio of 3%, the ITS and IT Mr of MNR-AC reached a highest value of 437.1 kPa and 3,896 MPa, respectively, while the lowest rut depth was 11.32 mm, which are significantly superior to those of conventional AC values. Furthermore, the MNR-AC demonstrated a highest ITFL of 1521, 912, and 523 pulses at 250, 300, and 350 kPa, respectively.

The performance of MNR-AC was found to be dependent upon elastic recovery, where the optimal r/b ratio for the best performance was found to align with the ideal ratio for the maximum elastic recovery. This suggests that improved elastic recovery enhances the adhesive and cohesive characteristics of the modified asphalt concrete, leading to overall performance improvements.

The relationship between the IT Mr and ITS of MNR-AC indicates a clear linear correlation across all r/b ratios. The analysis revealed that as ITS increased, the IT Mr also improved, reinforcing the conclusion that higher tensile strength directly enhances the material's elastic performance.

A novel fatigue distress model revealed a distinct logarithmic correlation between ITFL and initial tensile strain. The proposed model for conventional AC demonstrates a similar slope compared to the distress model for the standard asphalt concrete described by AI (1981) [33] but with different positions. This difference might be due to variations in material properties. When comparing the distress model of MNR-AC with that of conventional AC, there is an intersection between these two models. The intersection point is at an approximately 3–5% r/b ratio. This means that for the same applied stress, the MNR-AC with a r/b ratio \leq 3% has a higher ITFL than the conventional AC. Based on this developed distress model, the pavement design of MNR-AC can be conducted, which will promote the significant usage of MNR in sustainable pavement construction.

While MNR-AC has demonstrated significant improvements in both mechanical and fatigue properties, several limitations must be acknowledged. For instance, a comprehensive economic analysis was not conducted in this study. Future research should focus on evaluating the lifecycle costs and potential long-term maintenance savings to confirm the economic viability of MNR-AC.

To address these gaps, future research should broaden the scope by testing MNR-AC performance under a wider range of environmental conditions, including temperature variations and long-term aging effects. Field trials would provide a more accurate evaluation of real-world durability. Moreover, a detailed cost–benefit analysis comparing MNR-AC with other modifiers, including maintenance and lifespan considerations, is crucial. It is also important to explore the environmental sustainability of MNR-AC, particularly in terms of recycling, and its contribution to the circular economy in sustainable construction practices.

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References

- 1. Buritatum, A.; Suddeepong, A.; Horpibulsuk, S.; Akkharawongwhatthana, K.; Yaowarat, T.; Hoy, M.; Arulrajah, A. Improved performance of asphalt concretes using bottom ash as an alternative aggregate. *Sustainability* **2022**, *14*, 7033. [CrossRef]
- 2. Chen, J.S.; Liao, M.C.; Tsai, H.H. Evaluation and optimization of the engineering properties of polymer-modified asphalt. *Pract. Fail. Anal.* 2002, *2*, 75–83. [CrossRef]
- 3. Goodrich, J.L. Asphalt and polymer modified asphalt properties related to the performance of asphalt concrete mixes (with discussion). *Assoc. Asph. Paving Technol. Proc.* **1988**, *57*, 116–175.
- 4. Yildirim, Y. Polymer modified asphalt binders. Constr. Build. Mater. 2007, 21, 66–72. [CrossRef]
- Xing, C.; Tang, S.; Chang, Z.; Han, Z.; Li, H.; Zhu, B. A comprehensive review on the plant-mixed cold recycling technology of emulsified asphalt: Raw materials and factors affecting performances. *Constr. Build. Mater.* 2024, 439, 137344. [CrossRef]
- Kim, K.W.; Kweon, S.J.; Doh, Y.S.; Park, T.S. Fracture toughness of polymer-modified asphalt concrete at low temperatures. *Can. J. Civ. Eng.* 2003, *30*, 406–413. [CrossRef]
- Montanelli, E.F. Fiber/polymeric compound for high modulus polymer modified asphalt (PMA). *Procedia-Soc. Behav. Sci.* 2013, 104, 39–48. [CrossRef]
- 8. Takaikaew, T.; Tepsriha, P.; Horpibulsuk, S.; Hoy, M.; Kaloush, K.E.; Arulrajah, A. Performance of fiber-reinforced asphalt concretes with various asphalt binders in Thailand. *J. Mater. Civ. Eng.* **2018**, *30*, 04018193. [CrossRef]
- 9. Wang, P.Y.; Wen, Y.; Zhao, K.; Chong, D.; Wong, A.S. Evolution and locational variation of asphalt binder aging in long-life hot-mix asphalt pavements. *Constr. Build. Mater.* **2014**, *68*, 172–182. [CrossRef]
- Behnood, A.; Gharehveran, M.M. Morphology, rheology, and physical properties of polymer-modified asphalt binders. *Eur. Polym. J.* 2019, 112, 766–791. [CrossRef]
- 11. Kasikitwiwat, P.; Jantarachot, K. Comparative study of fatigue of asphalt concrete mixed with AC 60–70 and polymer modified asphalt binder. *Sci. Technol. Asia* 2021, *26*, 39–49.
- 12. Al-Mansob, R.A.; Ismail, A.; Yusoff, N.I.M.; Albrka, S.I.; Azhari, C.H.; Karim, M.R. Rheological characteristics of unaged and aged epoxidised natural rubber modified asphalt. *Constr. Build. Mater.* **2016**, *102*, 190–199. [CrossRef]
- Subagio, B.S.; Hariyadi, E.S.; Maha, I. The rutting resistance and resilient moduli of Pre-Vulcanized Liquid Natural Rubber modified asphaltic concrete in warm-mix temperature condition. J. Civ. Eng. Manag. 2022, 28, 196–207.
- 14. Muhammad, B.; Ismail, M.; Bhutta, M.A.R.; Abdul-Majid, Z. Influence of non-hydrocarbon substances on the compressive strength of natural rubber latex-modified concrete. *Constr. Build. Mater.* **2012**, *27*, 241–246. [CrossRef]
- 15. Al-Sabaeei, A.; Yussof, N.I.M.; Napiah, M.; Sutanto, M. A review of using natural rubber in the modification of bitumen and asphalt mixtures used for road construction. *J. Teknol.* **2019**, *81*. [CrossRef]
- Hoy, M.; Suddeepong, A.; Horpibulsuk, S.; Akkharawongthattana, K.; Arulrajah, A.; Buritatum, A.; Rashid, A.S.A. Improved Performance of Natural Rubber Latex–Modified Asphalt Concretes with Various Types of Aggregates. *J. Mater. Civ. Eng.* 2024, 36, 04023495. [CrossRef]
- Aiamsri, K.; Buritatum, A.; Suddeepong, A.; Horpibulsuk, S.; Yaowarat, T.; Hoy, M.; Phunpeng, V.; Arulrajah, A. Performance evaluation of pre-vulcanized rubber latex modified asphalt concrete with various aggregates. *Int. J. Pavement Eng.* 2024, 25, 2404705.
- 18. Wititanapanit, J.; Carvajal-Munoz, J.S.; Airey, G. Performance-related and rheological characterisation of natural rubber modified bitumen. *Constr. Build. Mater.* **2021**, *268*, 121058. [CrossRef]
- 19. Jitsangiam, P.; Nusit, K.; Phenrat, T.; Kumlai, S.; Pra-ai, S. An examination of natural rubber modified asphalt: Effects of rubber latex contents based on macro-and micro-observation analyses. *Constr. Build. Mater.* **2021**, *289*, 123158. [CrossRef]
- Al-Mansob, R.A.; Ismail, A.; Alduri, A.N.; Azhari, C.H.; Karim, M.R.; Yusoff, N.I.M. Physical and rheological properties of epoxidized natural rubber modified bitumens. *Constr. Build. Mater.* 2014, 63, 242–248. [CrossRef]
- 21. Suwarto, F.; Parry, T.; Thom, N.; Airey, G.; Abed, A.; Rahman, T.; Wititanapanit, J. Engineering characterization and environmental analysis of natural rubber latex modified asphalt mixture. *Constr. Build. Mater.* **2023**, 402, 132970. [CrossRef]
- Al-Mansob, R.A.; Katman, H.Y.; Al-Sabaeei, A.M.; Zamzami, M.; Al-Fakih, A.; Wei, W.K.; Koting, S. Effect of carbon nanofibers on physical, adhesion and rheological properties of liquid epoxidized natural rubber modified asphalt. *Materials* 2022, 15, 3870. [CrossRef] [PubMed]
- Udomchai, A.; Buritatum, A.; Suddeepong, A.; Horpibulsuk, S.; Yaowarat, T.; Akkharawongthattana, K.; Hoy, M.; Phunpeng, V.; Arulrajah, A.; Chinkulkijniwat, A. Asphalt concrete improved by concentrated rubber latex and prevulcanized rubber latex –Performance and cost analysis. *Case Stud. Constr. Mater.* 2024, 21, e03919. [CrossRef]
- 24. Tangboriboonrat, P.; Rakdee, C. Preparation of carbon black/natural rubber latex masterbatch in bead form. *J. Appl. Polym. Sci.* **2001**, *82*, 489–498. [CrossRef]
- 25. Xu, J.; Li, S.; Li, Y.; Ta, X. Preparation, morphology and properties of natural rubber/carbon black/multi-walled carbon nanotubes conductive composites. *J. Mater. Sci. Mater. Electron.* **2016**, *27*, 9531–9540. [CrossRef]
- 26. TIS 2731/2559; Natural Rubber Modified Asphalt Cement. Thailand Industrial Standard: Bangkok, Thailand, 2016.
- 27. DH-S 408/1989; Standard for Highway Construction. Thailand Department of Highways: Bangkok, Thailand, 1989.
- 28. DH-SP 401/1988; Specification for Asphalt Cement. Thailand Department of Highways: Bangkok, Thailand, 1988.
- 29. DH-T 604/1974; Standard for Asphalt Testing. Thailand Department of Highways: Bangkok, Thailand, 1974.

- 30. *ASTM D6931-17*; Standard Test Method for Indirect Tensile (IDT) Strength of Asphalt Mixtures. ASTM: West Conshohocken, PA, USA, 2017.
- 31. *ASTM D4123-82*; Standard Test Method for Indirect Tension Test for Resilient Modulus of Bituminous Mixtures. ASTM: West Conshohocken, PA, USA, 1995.
- 32. *EN 12697-24*; Bituminous 830 Mixtures-Test Methods for Hot Mix Asphalt-Part 24: Resistance to 831 fatigue. European Committee for Standardization: Brussels, Belgium, 2004.
- 33. *AASHTO T324*; Hamburg Wheel-Track Testing of Compacted Hot Mix Asphalt (HMA). Association of State Highway and Transportation Officials (AASHTO): Washington, DC, USA, 2014.
- 34. AI. Thickness Design-Asphalt Pavements for Highways and Streets; Asphalt Institute: Lexington, KY, USA, 1981; Volume 1.

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