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Mechanical Properties and Image-Based Crack Analysis of Hydrogenated Cup Lump Natural Rubber-Modified Asphalt Mixtures

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ABSTRACT

Researchers have explored the effects of incorporating cup lump natural rubber as an asphalt modification to promote rubber utilization and enhance pavement performance. However, cup-lump natural rubber treatment affects the performance of asphalt mixtures. This study investigates how cup-lump natural rubber treated via non-catalytic hydrogenation to create hydrogenated cup-lump natural rubber (HCLNR) affects the mechanical performance of asphalt mixtures. Asphalt mixtures were prepared by incorporating HCLNR at 2.5%, 5.0%, 7.5% and 10% by weight of bitumen. The performance was evaluated through Marshall stability, indirect tensile strength, and tensile strength ratio (TSR) tests, complemented by a MATLAB-based digital image analysis to quantify surface cracking and pattern. Results indicate that all HCLNR-modified mixtures comply with Malaysian Public Works Department specifications. The 5.0% HCLNR dosage was identified as the optimum content, achieving a peak Marshall stability of 13.1 kN and tensile strength of 862.08 kPa. Furthermore, the optimal dosage enhanced moisture resistance, with a TSR of 87.09% and the lowest crack ratio. Higher rubber contents increased bitumen stiffness and led to poor dispersion and brittle fracture patterns. This study confirms that HCLNR at moderate dosages serves as a sustainable and high-performance modifier, effectively improving the durability and crack resistance of asphalt pavements in tropical climates.

Keywords: Hydrogenated cup lump natural rubber; asphalt modification; Indirect Tensile Strength; Tensile Strength Ratio; crack analysis

INTRODUCTION

Asphalt mixtures are composite materials that are mainly applied in pavement engineering and other applications where durable and weather-resistant surfaces are required. Hot Mix Asphalt (HMA) is a commonly used construction material. It is black, viscous, sticky, and soluble in carbon disulfide. Polycyclic aromatic hydrocarbons are substances in HMA that make it flexible (Abdullah et al. 2014). But HMA is temperature-susceptible, which becomes soft at high temperatures and brittle at low temperatures. It also oxidizes rapidly in oxidative aging (Abdullah et al. 2014). Pavement distress is compounded by heavy traffic and overloading, especially in wet tropical climates. This is a combined effect and requires regular maintenance, raising operational costs. To ensure an adequate service life, therefore, it is important that strategies are developed to improve the performance of the asphalt and the overall durability of pavements. One approach is to modify bitumen using rubber.

Rubberized bitumens are gaining in use for road construction applications because they provide increased durability and better skid resistance (Othman et al. 2018). Latex improves performance by decreasing aggregate stripping and increasing resilient modulus (increases fatigue resistance and decreases permanent deformation). Ali et al. (2013) reported that the addition of natural rubber to the mixture resulted in an increase in the stiffness and rutting resistance of the mixture. A study by Krishnapriya (2015) confirmed that incorporating natural rubber into asphalt pavements enhanced rutting resistance and fatigue life. Shaffie et al. (2015 examined the stripping behavior and volumetric properties of asphalt mixtures containing natural rubber and granite aggregates. They concluded that the local aggregates used in their study fulfilled the Superpave consensus and source requirements. The enhanced stripping resistance on the pavement surface is due to the natural rubber polymer effectively coating the aggregates.

In order to stabilize rubber prices and lessen the impact on 300,000 smallholders, Malaysia made a smart move to enhance domestic consumption of natural rubber in asphalt for road pavement (Kamal et al. 2025). Hence, domestic demand for rubber is expected to rise due to the use of cup lump rubber in road construction. Cup-lump natural rubber (CLNR) offers a viable approach to improving pavement performance. CLNR is a solid by-product of the latex, typically cup-shaped lumps, collected from rubber trees (Rohayzi et al. 2023). The Malaysian Rubber Board (MRB) first introduced cup-lump rubber as an additive in asphalt mixtures to address the declining price of natural rubber, particularly cup-lump rubber. Recent studies have demonstrated the potential of CLNR as a sustainable modifier that enhances asphalt characteristics and performance. They reported that the addition of CLNR increases the softening points and rutting resistance (Azahar et al. 2019). In addition, CLNR is characterized by superior fatigue resistance, high stretch ratios, and excellent moisture durability (Ikeda et al., 2018). These attributes, combined with its cost-effectiveness and environmental benefits, have supported its use in road construction for several decades (Al-Sabaeei et al. 2019; Ansari et al. 2022). CLNR modification has been formally adopted by the Malaysian Public Works Department (PWD) for pavement applications (Ghafar et al. 2022).

Previous studies reported higher resilient modulus and improved dynamic creep behavior of asphalt mixtures containing CLNR, which are the key indicators of tensile strength under repeated loading (Azahar et al. 2019; Zheng et al. 2024). The rubber particles in CLNR enhanced the elasticity of asphalt mixtures, thereby increasing rutting resistance. Shaffie et al. (2017) found that modified mixtures have resulted 41% lower rut depth than the unmodified sample, indicating the effectiveness of CLNR in mitigating permanent deformation. Researchers investigating the viscoelastic response of CLNR-modified asphalt found that its rubber content is critical in enhancing the bitumen's tensile load capacity (Ansari et al. 2021; Azahar et al. 2019). Studies by Krishnapriya (2015), Shaffie et al. (2015), and Tuntiworawit et al. (2005) found that rubberized asphalt mixtures improve tensile strength more than conventional mixtures. The higher strength indicates the better ability of rubberized mixtures to withstand higher strains before failure under static loading at intermediate temperatures.

A case study was done by Othman et al. (2018) at Jalan Kuala Lumpur-Kuantan reported that the integration of 5% cup lump modified bitumen (CMB) into the cup lump modified asphalt (CMA) mixture yielded a performance profile that satisfies PWD requirements, with significant enhancements over conventional AC14 pavement. The improved performance of the CMA pavement is demonstrated in the very good structural condition (100%) and good surface characteristics, such as skid resistance and resistance to permanent deformation. Razali et al. (2019) have found that the use of CMA mixtures can enhance pavement performance without requiring major changes to current construction techniques. Throughout a six-month testing period. Comparative site tests showed that the CMA pavement performed on a level equal to conventional asphalt.

Although the use of natural rubber in asphalt is beneficial, previous studies on the use of natural rubber for bitumen modification have identified several issues. The effect of rubber and bituminous interaction during blending on the physical characteristics of natural rubber and its performance is explained by Kintali and Anirudh (2015). Conventional solvent-based pretreatment, such as soaking in toluene, is often used to enhance the rubber interaction and to obtain a homogeneous blend. Toluene helps disperse the rubber but can cause bitumen to age if mixed for a long time, and it poses safety concerns because it is highly flammable.

Therefore, this study introduces a new chemical modification method to overcome the drawback of CLNR bitumen modification, which is through hydrogenation. The process begins with a pre-treatment of CLNR to modify its physical structure, then a non-catalytic hydrogenation process is carried out using diimide as a hydrogen source to obtain a hydrogenated cup lump natural rubber (HCLNR). Although this modified bitumen shows strong potential, its influence on important asphalt performance properties such as stiffness, resistance to moisture, and cracking behaviour has not yet been thoroughly studied. To address this gap, this study investigates the mechanical performance of the HCLNR-modified mixture using Marshall properties, indirect tensile strength, moisture resistance, and crack-behavior analysis.

METHODOLOGY

MATERIALS AND SAMPLE PREPARATION

The cup lump natural rubber (CLNR) was obtained from a local plantation in Telipok, Sabah, and the 60/70 penetration grade bitumen was supplied by Astenpave Sdn. Bhd. The CLNR was pretreated by soaking in toluene at a 1:5 CLNR-to-toluene ratio for two weeks with periodic stirring, followed by hydrogenation to improve compatibility. The procedure of hydrogenation was adopted by Jamaluddin et al. (2016). The carbon–carbon double bonds in the CLNR were hydrogenated in a non-catalytic process using diimide to release the hydrogen molecules. The diimide was synthesized from p-toluenesulfonyl hydrazide (TSH). It was added to the treated CLNR solution at a 1:1 weight ratio (TSH: CLNR). Then, the mixture was heated to 130 °C under constant stirring to generate HCLNR. The resulting hydrogenated CLNR was blended with bitumen using a mechanical mixer at 160 °C for 30 minutes at 1000 rpm. In this study, HCLNR was incorporated as a bitumen replacement at percentages of 2.5%, 5.0%, 7.5%, and 10% by weight of the bitumen. The selection of these dosages is consistent with the 2.5% incremental approach employed by Abdulrahman et al. (2019) and Albuaymi et al. (2023). The maximum content of 10% was determined according to previous studies, which showed that rubber content higher than 9% was generally not compatible with the standard storage stability requirements, as it was determined that high molecular weight natural rubber causes instability in rubber (Al-Mansob et al. 2014;Yıldırım 2007). In addition, when the rubber content exceeds 10%, the rubber content may cause certain particles to remain undissolved, and the bitumen may become non-homogeneous, which affects the storage stability and mechanical properties of the rubberized asphalt (Ansari et al. 2022).

For the asphalt mix design, it was adopted from a local premix supplier that supplies aggregate for this study. The aggregates gradation followed the ACW20 gradation of PWD Malaysia (Table 1), with an optimum bitumen content of 5.47%. Mixing and compaction temperatures were maintained at 160 °C and 150 °C, respectively. The physical properties of the conventional and HCLNR-modified bitumen were assessed and benchmarked against PWD Malaysia specifications, as presented in Table 2. From the table, it can be seen that hydrogenated CLNR hardens the asphalt bitumen with lower penetration and high ductility compared to the conventional asphalt. The softening point was marginally below the specified limit; however, the deviation was minimal and remained close to the target range. This indicates general conformity with the specification, with only a slight potential for increased mixture flow under local temperature conditions (Steven et al. 2023).

TABLE 1. Aggregate gradation ACW 20

Sieve Size (mm)	Percentage Passing (%)
28	100
20	76 -100
14	64 - 89
10	56 - 84
5	46 - 71
3.35	32 - 58
1.18	20 - 42
0.425	12 - 28
0.15	6 - 16
0.075	4 - 8

TABLE 2. Physical properties of the conventional bitumen and hydrogenated cup lump natural rubber (HCLNR)

Properties	CB	HCLNR (%)			
		2.5	5.0	7.5	10.0
Penetration at 25°C (dmm)	63	60	60	70	72
Softening point (°C)	45	46	47	44	45
Viscosity at 135°C (Pa.s)	0.63	0.79	0.81	1.18	1.4
Ductility at 25°C (cm)	130	125	134	131	116

EXPERIMENTAL DESIGN AND PROCEDURE

The mechanical performance of asphalt mixtures with varying HCLNR percentages was evaluated through a series of standardized tests. It is used to determine the maximum load of compacted asphalt mixture that could withstand, tensile resistance, moisture susceptibility, and cracking behavior of the asphalt mixtures as specified by ASTM standards. The Marshall stability and flow test (ASTM D6927, 2015) is used to ensure asphalt pavements can endure heavy traffic without permanent damage. Indirect tensile strength (ITS) test (ASTM D6931, 2012) was performed to determine their crack resistance under traffic loading and environmental stresses. Moisture susceptibility was determined through the tensile strength ratio (TSR) test, following ASTM D4867. The specimens were conditioned by immersing them in 60 °C water for 24 hours, and comparing the ITS with the unconditioned samples. To ensure the reliability and reproducibility of the experimental results, all mechanical and physical tests were conducted in triplicate (three independent specimens per mixture type).

Afterward, the MATLAB-based digital image method was used to quantitatively evaluate surface damage and identify crack patterns, which were adapted from Hoang et al. (2018). While Hoang utilized convolutional neural networks (CNNs) for classification, this study focused on a streamlined MATLAB-based processing workflow to calculate crack ratios and infer mechanical behavior (e.g., stiffness and fatigue susceptibility). The crack analysis workflow is shown in Figure 1, whereby cracked specimen images were imported into MATLAB software and converted to grayscale to remove color variations. Canny edge detection has then been used to enhance the crack boundaries with high accuracy. The edge-detected images were subsequently converted into a binary format, isolating the cracks as white pixels against a black background. The amount of damage was measured using the "crack ratio", which is the number of white pixels divided by the total number of pixels. Finally, the binary images were qualitatively evaluated with respect to the morphology of the cracks, including their shape, orientation, and continuity of block and longitudinal cracks.

RESULTS AND DISCUSSION

MARSHALL STABILITY AND FLOW

The Marshall stability and flow results for HCLNR-modified asphalt are shown in Figure 2. The stability of the control bitumen (CB) was 13.67 kN, which was higher than the minimum requirement of PWD of 13.0 kN. Initial modification with 2.5% HCLNR reduced the stability to 11.06 kN, while 5% HCLNR (13.10 kN) met the PWD requirement and was found to be the optimum level of interaction between the rubber and the bitumen matrix. The dosages of 7.5% and 10% exhibited a downward curve, as did all other dosages, following the typical bell-shaped performance curve.

As for the flow values, all of the mixtures were within the limits recommended by the PWD of 2.0–4.0 mm. The 2.5% and 5% dosages had a more balanced flexibility, while the 7.5% HCLNR mixture had the lowest flow (2.65 mm), indicating that the mixture was stiffer and may be more prone to cracking. On the other hand, the highest flow (3.85 mm) was obtained from the 10% HCLNR content and may cause rutting under heavy traffic loads. Overall, the 5% HCLNR dosage shows improvements in mechanical performance, aligning with findings by Ali et al. (2013) and Krishnapriya (2015).

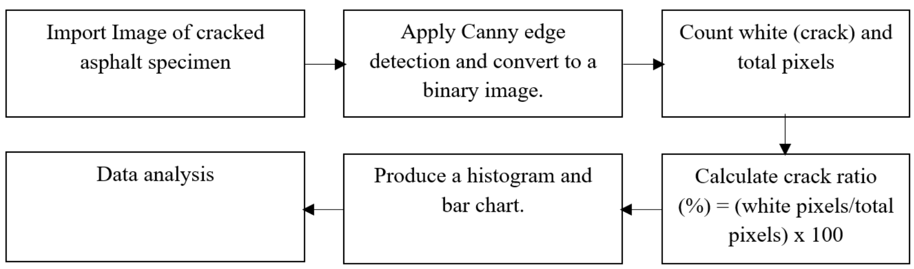

FIGURE 1. Workflow of crack analysis

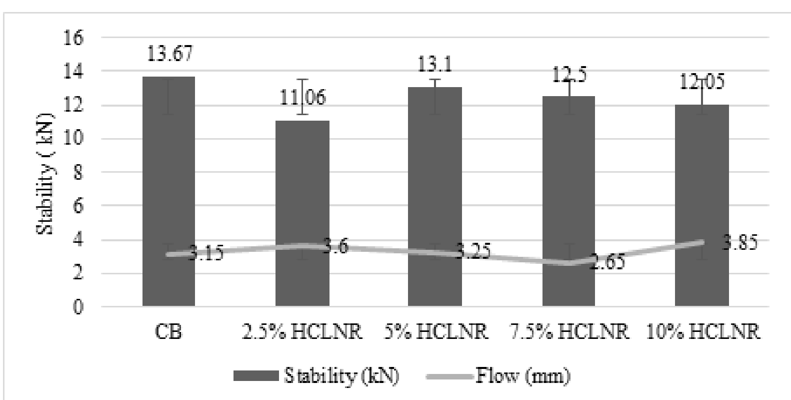

FIGURE 2. Stability and flow of the HCLNR modified asphalt mixtures

INDIRECT TENSILE STRENGTH

Figure 3 presents the ITS values for the control bitumen (0% HCLNR) and those modified with different HCLNR percentages. The inclusion of HCLNR influenced the tensile strength, whereby 5% HCLNR was the highest among the HLNCR content. Incorporating 5% HCLNR increased the ITS to 862.08 kPa, which is slightly higher than the control bitumen, indicating better bitumen elasticity and aggregate–bitumen cohesion. The ITS result at 2.5% may not accurately reflect the material behavior and could be attributed to potential experimental variability. Furthermore, the literature indicates that most studies on cup lump natural rubber have focused on contents above 5.0%. The increased on ITS at 5.0% is aligned with those for the unmodified and modified bitumen in a study conducted by Shaffie et al. (2015) in which mixtures containing natural rubber consistently exhibited higher indirect tensile strength. The HCLNR-modified rubberised and polymer-modified asphalt mixtures obtained in this study have an ITS within the normal range reported for these mixtures, indicating similar mechanical performance. According to the literature, the ITS of cup lump natural rubber-modified mixtures typically ranges from 700kPa to 950 kPa (Ansari et al. 2023; Azahar et al. 2019). Ansari et al. (2023) reported that 6% CLNR dosage yields an ITS value of 900kPa. This performance threshold is consistent with the findings of this study, in which a 5% HCLNR mixture achieved a peak strength of 862.08kPa. The differences observed can be explained by differences in aggregate gradation, bitumen characteristics, and mix design parameters.

However, increasing the HCLNR percentages to 7.5% and 10% reduced the tensile strength to 801.94 kPa and 761.84 kPa, respectively. This could be the result of high HCLNR, which could not disperse evenly, leading to some weak areas and inconsistencies. A recent study by Putri et al. (2025) supports this, confirming that a higher modifier content leads to over-saturation, which creates weak spots and decreases effective binding between aggregates. This behavioral trend is also consistent with findings by Azahar et al. (2019) who explicitly noted that incorporating natural rubber at concentrations exceeding 9% compromises the physical attributes of the bitumen, yielding a low-quality rubberized asphalt compared to conventional bitumen. Results from the Mohammed Aboud et al. (2020) indicate that a progressive increase in tensile strength was observed as the polymer concentration rose, peaking at a 6% threshold before declining at the 8% dosage level. The modified mixtures consistently exhibited significantly higher tensile capacity compared to the control. This performance enhancement is attributed to the increased structural stiffness due to polymer modification.

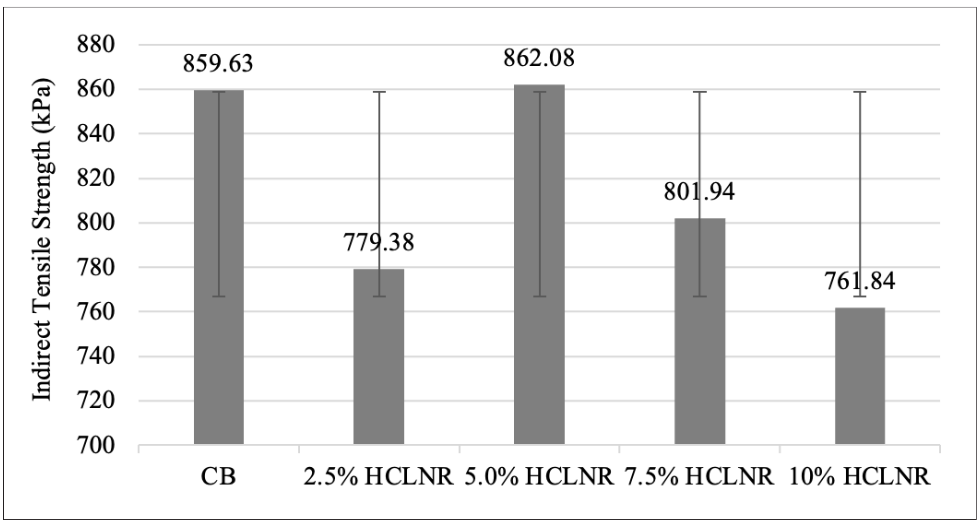

FIGURE 3. ITS of the HCLNR modified asphalt mixtures

Moisture damage contributes to weakening the hot-mix asphalt's bitumen–aggregate adhesion. The tensile strength ratio measures the tensile strength before and after conditioning. It provides a reliable measure of the asphalt mixture's water sensitivity. A minimum TSR of 80% is a generally accepted value for field performance (JKR, 2020). Figure 4 presents the TSR results for the mixture with different HCLNR percentages. The control mixture's TSR of 83.73% meets the standard threshold. Incorporating 2.5% HCLNR increased the TSR value slightly to 85.33%. The highest TSR value of 87.09% was achieved with 5% HCLNR, indicating enhanced cohesion and stripping resistance under wet conditions. This finding aligns with an earlier study reporting higher TSRs (Mamhuseesini and Nareeman, 2018), indicating enhanced water resistance, as mixtures with higher values exhibit better tensile strength retention under wet conditions.

Incorporating higher HCLNR percentages of 7.5 and 10% reduced the TSR values to 85.05% and 84.28%, respectively. However, these values are still above the minimum requirement. The lower TSR values are attributed to poor dispersion of the excessive rubber, leading to the formation of rubber-rich zones that compromise uniform stress transfer (Cao et al., 2010).

In general, all the HCLNR mixtures have a TSR value above 84%, which means there is an improvement in moisture resistance compared to the control specimen. The best performance was obtained with 5% HCLNR, which is in line with the findings of Krishnapriya (2015) and Mamhuseesini and Nareeman (2018). The mixture with the concentration of 5% HCLNR exhibited an optimal balance of adhesion, cohesion, and workability. The findings of this study are very significant to areas with tropical support for the potential of HCLNR as a sustainable and durable asphalt modifier.

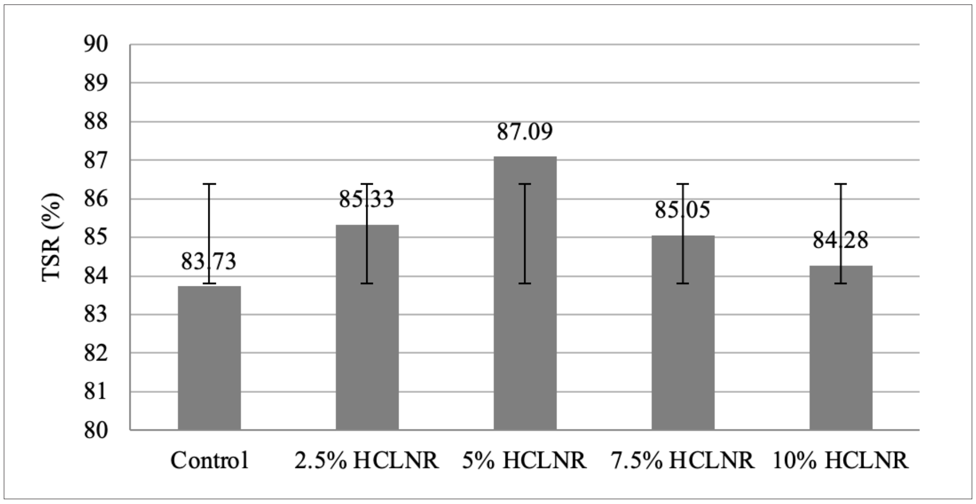

FIGURE 4. Effects of adding HCLNR on TSR values

IMAGE-BASED CRACK ANALYSIS

The specimens tested using indirect tensile strength (ITS) were photographed following failure. The images were processed using MATLAB, and the severity of the cracks was quantified based on the pixel intensity. This was done to enable the evaluation of the cracking behavior of mixtures in a consistent and non-destructive method, and to understand the role of the modifier in improving fracture resistance, with the mixes composed of 0%, 2.5%, 5%, 7.5%, and 10% HCLNR. The image-based analysis quantified crack propagation by converting the specimen images to grayscale and by using a threshold intensity of 127 to distinguish crack pixels from intact surfaces. The crack ratio was determined as the percentage of crack pixels in the 100 mm circular area of the specimen minus the background area.

Quantitative analysis showed a non-linear relationship between the HCLNR percentage and the crack ratio (Table 3). The control mix (0% HCLNR) has the highest crack ratio (33.36%), consistent with the ITS and TSR values, indicating poor flexibility and high susceptibility to tensile failure. Although incorporating 2.5% HCLNR reduced the crack ratio to 26.24%, the specimen still exhibited block-like cracking. Optimal performance was achieved with 5% HCLNR, with a crack ratio of 19.23%. The crack is a single narrow longitudinal crack, suggesting enhanced ductility and resistance to tensile stress. Incorporating more than 5% HCLNR resulted in higher crack severity. The mixture with 7.5% HCLNR exhibited branched cracking (22.75%), and that with 10% HCLNR showed severe multi-directional cracks (29.17%), indicating an over-stiffened mixture and reduced strain accommodation.

TABLE 3. Crack ratio with HCLNR content

HCLNR (%)	Crack Ratio (%)
0	33.36
2.5	26.24
5.0	19.23
7.5	22.75
10	29.17

Generally, the transition from widespread block cracking in the control mix to localized longitudinal cracks with the incorporation of 5% HCLNR, and subsequently to brittle branched cracking at higher HCLNR percentages demonstrated the significant effect of modifier content on fracture performance (Figure 5). This study generated three types of images for each asphalt specimen to evaluate cracking behavior. The top image, in grayscale, is the original specimen surface. The binary image in the middle was produced by converting the pixels into black and white to distinguish cracks from intact regions. The overlay image (bottom) superimposes the detected cracks on the grayscale background. It facilitates a better interpretation of crack distribution on the surface texture. The binary image was used in quantitative crack analysis because it allows precise measurement of the crack area and distribution.

The initial visual inspection of crack patterns of the broken samples revealed that, in all cases, the cracks spread primarily through the mixture instead of through the aggregate particles. This consistent trend indicates that the mixture's mechanical properties, not its aggregate strength, govern fracture resistance. This result supports the well-established understanding that the elasticity and cohesion of flexible pavements are the critical factors resisting tensile stress and crack development. They confirm that incorporating 5% HCLNR enhances crack resistance, but excessive amounts of HCLNR lead to brittleness and crack propagation.

The lower crack ratio of 26.24% confirmed that incorporating 2.5% HCLNR enhanced the mixture's elasticity. The optimal performance from incorporating 5% HCLNR, as indicated by the lowest crack ratio (19.23%), suggests enhanced stress distribution and delayed crack initiation. Azahar et al. (2019) and Rohayzi et al. (2023) reported similar improvements in viscoelasticity and crack resistance at moderate rubber contents. However, higher HCLNR percentages lead to more severe cracking, with crack ratios of 22.75% (7.5%) and 29.17% (10%), which then disrupts the mixture's continuity.

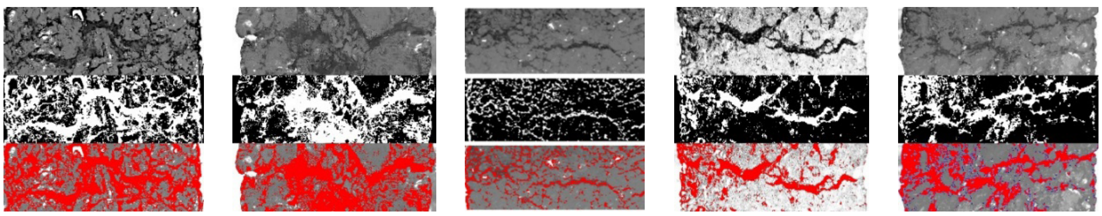

FIGURE 5. Grayscale (top), binary (middle), and overlay (bottom) crack images of asphalt specimens after ITS failure

The effect of HCLNR on asphalt performance does not exhibit a consistent trend with increasing HCLNR content. The asphalt mixture with the highest ITS (862.08 kPa) and TSR (87.09%) and the lowest crack ratio (19.23%) and simple longitudinal cracking pattern was the 5% HCLNR mixture. While having acceptable strength, the mixtures with higher HCLNR percentages (7.5% and 10%) have lower ITS and TSR values, and larger, branched cracking. Image analysis confirmed that cracks propagated primarily through mixture-rich zones and not through the aggregates. From these findings, it can be seen that the incorporation of a moderate amount of HCLNR resulted in improvement of tensile strength and moisture resistance and a decrease in the level of cracking at the surface. Conversely, using more HCLNR in the asphalt mix can lead to a stiffer and more brittle mixture with poorer cracking properties.

CONCLUSION

This study was done to assess the mechanical properties and cracking response of asphalt mixtures incorporating hydrogenated cup lump natural rubber (HCLNR). The laboratory test results indicated that mechanical properties were improved significantly when the HCLNR was added to the asphalt mixtures compared to when the HCLNR was not added (0% HCLNR). In particular, the content of 5.0% HCLNR met the requirement with a stability of 13.10 kN, close to the control mixture with 13.67 kN. In addition, the moisture susceptibility test showed that the moisture resistance of the 5.0% HCLNR mixture was best with a TSR of 87.09%. The optimum modifier content was determined to be 5% HCLNR. At this level, hydrogenation completed the saturation of the rubber backbone, which allowed the creation of a homogeneous polymer-bitumen network that improved the cohesion of aggregate-bitumen and bitumen elasticity. On the other hand, when the dosage was raised to 7.5% and 10%, the ITS and stability decreased due to the high amount of HCLNR, which was unable to disperse evenly, allowing for some weak points and variations. This is consistent with the results of the digital image analysis, in which the crack ratio was lowered to 19.23% and was the most resistant to surface damage.

Further studies are needed to validate the resulting improvements in field conditions and to evaluate the long-term performance of the resulting HCLNR-modified mixtures in a variety of climatic and traffic conditions. To further enhance the accuracy and efficiency of crack detection, the digital image analysis method developed in this study can be optimized using state-of-the-art computation tools, including machine learning algorithms or automated segmentation systems. Such enhancements would enable more accurate surface distress to be quantified and improve the reliability of image-based crack evaluations.

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DECLARATION OF COMPETING INTEREST

None.

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