Review on the potentials of natural rubber in bitumen modification

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Abstract. In recent times both natural and synthetic rubber were blended with bitumen with the hope of producing a more durable hot mix asphalt. Although the application of natural rubber (NR) in bitumen modification have started long time ago prior to crumb rubber; only few literatures that completely characterised its performance are found today. This review aims to remind researchers and paving agencies on the benefits of NR rubberised asphalt. The study draw attention to NR production process and its unique properties, how NR was used to solve four major asphalt pavement failures namely rutting, fatigue, thermal cracking, and moisture susceptibility. It also presents barriers limiting the application of NR in bitumen modification with possible solution. Lastly, the concept of applying NR in warm mix asphalt was introduced.

1. Introduction

Hot mix asphalt (HMA) is a combined mix of aggregate (about 94 - 96%) and asphalt binder (4 – 6%) by weight of the total mix. Although the percentage of bitumen is comparatively small; it significantly determines the performance and durability of the pavement more than the aggregate. This is because climatic factors such as heat, rain and sun radiation have more effect on the asphalt binder than on aggregate [1,2]. In the past, the use of unmodified bitumen in road construction was enough in supporting load from traffic flow. However, high traffic load placed on the road network nowadays has reached a critical stage. This is due to rapid increase in volume of heavy vehicles coupled with significant increase in allowable axle weight which leads to premature failure of flexible pavement [3]. In addition, HMA roads show susceptibility to temperature through softening at high temperature and cracking at low temperature. Therefore, it is essential to improve the quality of asphalt binder with a material that can lessen its temperature susceptibility, enrich cohesion, increase its viscosity and elasticity [4].

Through research the idea of modifying bitumen with polymer to increase the pavement design life and reduce frequent maintenance was developed. Polymer improves the rheological property of bitumen by dissolving and distribution its long chain molecules to create a network of inter-connecting polymers. This inter-connecting matrix of long chain polymer molecules is what improves the physical properties of bitumen. It is worthy to mention that the addition of polymer does not chemically alter the nature of the modified binder; but rather they only improve its physical properties [5,6]. The advantage of using polymer modified bitumen (PMB) include higher elastic recovery, higher softening point, greater viscosity, greater cohesive strength and ductility [7]. Thus, PMB has proven to be an indispensable part of pavement construction process [8,9]. Examples include polybutadiene, polyisoprene butadiene styrene (SBS), ethylene vinyl acetate (EVA), polyvinyl acetate (PVA), styrene butadiene rubber (SBR) and natural rubber (NR) [10,11].

Rubber is polymeric material endowed with the properties of flexibility and extensibility. When force is applied the molecules stretches out in the direction of the applied force, on release from extension they suddenly recover to their initial state. NR is a renewable and ecologically friendly elastomer which possesses good elasticity and outstanding properties under cyclical loads. It is more cost-effective compared to synthetic polymer modifiers for asphalt especially in the rubber producing countries [12,13]. The application of NR in bitumen modification have been used for years. In fact, the addition of NR in bitumen was patented in 1840 in England, and by 1920 Great Britain and United states have extensively tried to produce rubberised roads without asphalt. Over the last six decades crumb rubber was extensively studied by researchers around the world mainly because of its abundance as a waste product [14,15], and there is enough literatures on how crumb rubber improves the performance of HMA [16–19].

Although the application of NR in bitumen modification have started long before other polymer types, only few literatures that completely characterised its performance are found today. A decade ago, limited studies on advanced properties of NR modified bitumen mixes was pointed out as the major barrier limiting the adoption of NR modified asphalt in many places including the major rubber producing countries. Another reason is because few synthetic polymers such as SBS, EVA, SBR and polyolefin are widely produced and marketed at the expense of a renewable and sustainable resources like NR [20]. Sadly today, despite the advancement of technology and availability of advance testing equipment only few literatures that fully characterised the performance of NR modified asphalt are found. This is because researchers are more focused on incorporating waste materials such as waste engine oil, crumb rubber, waste cooking oil, palm oil fuel ash and coconut ash in bitumen modification [21–23]. This study was carried out to enlightening researchers and road agencies on the various benefit of incorporating NR in road construction. The review became necessary due to limited number of studies on the performance of NR bitumen at different temperature, durability, field trials, or in warm mix asphalt thus limiting its application into practice.

2. Natural rubber

NR occurs in almost 2000 plant species but only a few are industrially important. The first place being occupied by Hevea brasiliensis, a tree growing up to 30 m high with a 50 cm diameter of the main trunk. NR is an elastomer that was originally derived from milky latex found in the sap of some plants [24]. It is not advised to directly add the raw latex to bitumen because of its high-water content and defencelessness to bacteria attack. Methods commonly used in achieving high concentration of the solid rubber includes centrifugation, creaming and evaporation [25,26]. The current global production of NR is about 12.31 M tons amounting to 46% of the world's total rubber production. With Thailand as the world's biggest producer and exporter (37%) while China is the major consumer of the commodity [27–29].

2.1. NR production process

NR is extracted by tapping the bark of Hevea brasiliensis tree, the rubber latex is contained in cells which are formed by metabolic processes occurring in plants. The sticky liquid, of milky appearance that flows from the tree is a colloidal dispersion of polyisoprene molecules in an aqueous medium [10,30]. Figure 1 below shows a typical rubber taping process.

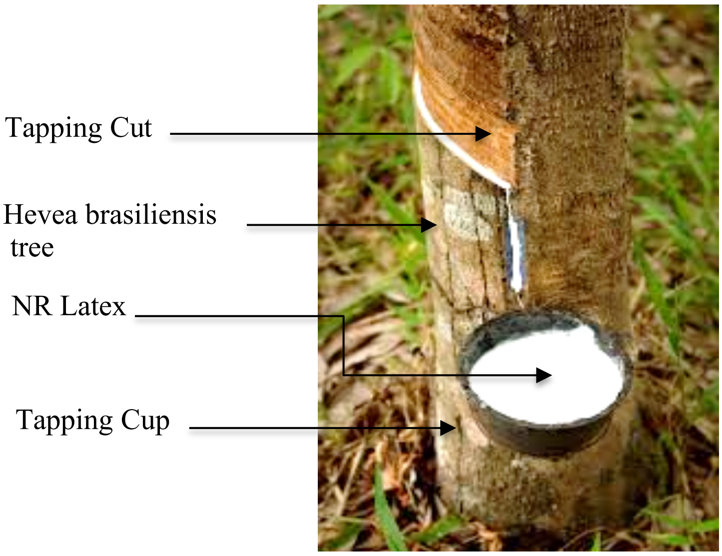


Figure 1. Freshly tapped NR latex [31].

To preserve the NR latex in its liquid form; alkaline solution (usually ammonia) is added to prevent its coagulation [32]. However, when the latex is allowed to remain in the tapping cup for a day or more, bacteria is produced which kickstart the coagulation process by removing excess water from the latex. The produced bacteria release positively charged ions which react with the negatively charged ions at the surface of the NR latex to initiate the coagulation process, this coagulated rubber is often referred to as "cuplump rubber". Tuntiworawit et al. (2005) defines cuplump rubber as a freshly coagulated rubber where the coagulation takes place in the cup at the tree [33]. To speed up the coagulation process, formic acid is usually added to the NR latex and the whole process is completed within 2 – 5 hours. NR latex particles was hypothesized to have a micellar structure (Figure 2) with hydrophobic rubber core surrounded by mix of lipids and protein at the surface [30].

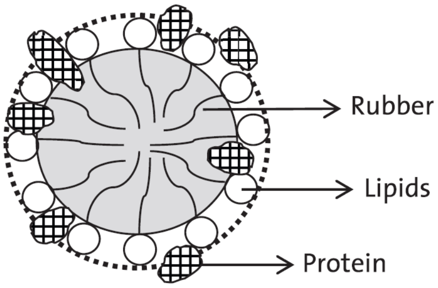
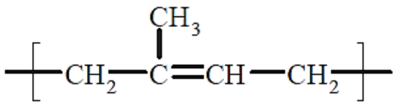


Figure 2. Cross-section of NR latex particle [30].

2.2. *Constituents of NR*

NR latex contains two phases namely the disperse phase and dispersion medium, the disperse phase consists of large number of small polymeric particles. The solid content of the latex is between 30 to 40% although it may rise to 45−50% after a long period of non-tapping. The rubber produced from latex contains hydrocarbon and relatively small quantities of protein, carbohydrates, resin-like materials, mineral salts and fatty acids [10,30]. The empirical formula of NR is $(C_5H_8)_n$ while the chemical structure composition is shown in Figure 3 [34,35]. It is imperative to note that the presence of C=C double bond made it feasible to chemically modify the properties of NR for certain applications [36].



$$\left[-CH_2-\overset{CH_3}{\overset{|}{C}}=CH-CH_2- \right]$$

Figure 3. Chemical structure of NR [37].

Table 1. NR composition [38–40].

Content	Value
Total solid content	≥ 61.50%
Dry rubber content	≥ 60.09%
Non-rubber content	≤ 2.00%
Ammonia content (on total weight)	0.65 – 0.75%
pH value	≤ 11.00
Potassium hydroxide number (KOH)	0.85
Maximum volatile fatty acid number (VFA)	≤ 0 .05
Mechanical stability time (seconds)	1227 (Good)
Specific gravity at 25°C	≥ 0.94
Magnesium (ppm)	29
Manganese content	Traces

2.3. Effect of shear rate and temperature on rheology of NR

Elastomeric polymers are flexible enough to be extended up to ten times without breaking and swiftly return to their original structure once the applied load is released. From rheological perspective, latex behaves like a complex colloidal suspension of polymeric substance in an aqueous medium [41,42]. Varkey et al., extensively studied the effect of shear rate on the rheology of NR latex [35,43]. The study found that NR exhibited a shear thinning behaviour (pseudoplastic) where increase in shear rate reduces the viscosity. Normally, links are formed when rubber particles in dispersion make contact, but under the influence of shearing stress the links are stretched, distorted, and ruptured leading to the reduction in viscosity. Similar studies by Lim et al. [44] and Bauer et al., [42] confirmed the above findings. Similarly, viscosity was found to decrease with an increase in temperature. However, at low shear rate the viscosities of the blends increase with increase in temperature. This is because new links are formed when the particles in dispersion make contact. Thus, the reason why asphalt-rubber blend needs to be mixed at high shear rate before a workable and homogeneous blend of the mix is achieved.

The ability of a material to maintain its strength, elasticity, toughness at a given or range of temperatures is referred to as thermal stability. A clear knowledge of how NR breakdown on heating is of paramount importance in determining the road performance of PMB. Varkey et al., (2000) found that the degradation of NR takes place in the region of 310 °C to 456 °C. It was observed that NR shows extensive weight loss of 91.59% and cross-linking at 300 – 450°C due to chain scission. Another study by Tinavallie [46] found that pure NR decomposed at 300 – 450 °C. In comparison; styrene butadiene rubber (SBR) was found to be more stable than NR because its degradation takes place around 390 – 520 °C and a weight loss of 85.57%. The study conclude that NR has less temperature susceptibility thus making it a better polymer for bitumen modification.

Similarly, because of NR's structural regularity it tends to crystallize spontaneously at lower temperatures. Although the low temperature crystallization causes stiffening, it can be easily reversed by warming. It is this strain-induced crystallization that gives products made from NR superior resilience, elasticity, tensile strength, low heat build-up and abrasion. These are some of the properties that makes the tyre industry to heavily depend on NR because its elastic properties remain unequalled to date [10,47].

3. NR resistance to major failure of asphalt

HMA performance is the resistance of asphalt binder to the combined destruction of heat, weather and time. Due to asphalt binder's susceptibility to temperature variation it becomes stiff at low temperatures and tend to crack under stress. For NR modified bitumen; the rubber acts like an elastic band in holding the bitumen together and dissipate the applied stress as it develops. Upon increase in temperature the bitumen deforms into a viscous liquid and begins to flow. But the presence of NR helps in resisting the flow by acting like a membrane [20].

3.1. Rutting

Rutting is a common form of distress on flexible pavement caused by increase in traffic volumes, truck loadings and higher tire pressures. This distress often leads to hydroplaning which endangers the road user' life. It also significantly increase the cost of roads maintenance, fuel consumption as well as reduce the service life of the pavement [7,48,49]. A study by Sai & Gottala [50] found that addition of NR decreases the penetration value and increases the softening point of 80/100 PEN bitumen and conclude that the resistance to flow of the bitumen increases with increase in the percentage of NR. Similarly, NR in the form of ribbed smoked sheet was added to a 60/70 grade bitumen. It was mixed using high shear mixer at 150 - 170 °C for 2 hours, then incubated at 120 °C for 1 day. The study found that the addition of NR results in the asphalt binder to have lower penetration value with high softening point, penetration index, torsional recovery and tenacity [4]. This indicates that NR-modified asphalt paved roads shall have more strength and durability than the unmodified binder.

Furthermore, the rutting performance of incorporating NR Latex modified binder on the engineering properties of SMA and dense graded mixtures was carried out by Shaffie et al., [7]. The result show that the modified mix have better resistance to rutting than the conventional HMA mix and conclude that SMA14 mixture exhibits better rutting resistance than the dense graded AC14 by 41%. Also a recent research by Siswanto [51] on addition of 6% NR latex to a 60/70 Pen bitumen significantly reduced the permanent deformation of the asphalt mix from 9.73 mm to 6.20 mm (36%). And the following conclusions were made, NR reduced the rate of deformation by increasing the dynamic stability of asphalt mixes. Also, the rate of deformation resistance is directly proportional to the amount of NR added. Lastly, Krishnapriya [6] also found that NR modified bituminous mixes exhibits excellent rut resistance.

3.2. Fatigue cracking

One of the most common forms of cracking on asphalt pavement is fatigue cracking. It occurs from accumulated strain on the pavement binder induced by repeated traffic loading. The peak tensile strain occurs at the bottom layer and once the crack is initiated, it propagates upwards and weakened the whole pavement structure. Factors responsible for this type of cracking include heavy traffic loading, resilient modulus of the material, subgrade drainage, and adhesive failure of the underlying pavement layers [52] [6].

A study was carried to evaluate the performance of NR modified asphalt and to determine the effect of aggregate gradation at lower limit, middle limit and upper limit of the mix. The outcome shows that the percentage of air void has substantial impact on fatigue life of the mix. This is because the fatigue life of the asphalt binder decreases with increasing percentage of air void. Furthermore, the fatigue life of the asphalt binder significantly increased when the rest period of 0.9 seconds was introduced. Resting period allows the HMA to gain some strength before subjecting it to repeated cyclic loadings, thus simulating field conditions at the laboratory. Lastly the middle aggregate gradation has the best performance values when compared with the lower and upper gradations [6].

3.3. Stripping failure

Stripping is the physical separation of the bitumen from the aggregate due to loss of bond between the asphalt binder and aggregate caused by the action of water. Factors responsible for moisture susceptibility of asphalt mixes include type of mix, asphalt binder, aggregate characteristics, and the pavement environmental conditions during and after construction [53–55].

Shaffie et al., [55] used Lottman and boiling water test to assess the stripping performance of NR modified bitumen. The result shows that the tensile strength values are greater for NR modified mix compared to the unmodified binder, indicating better resistance to stripping. The study concludes that addition of NR to bitumen significantly improves the cohesion and adhesion property of a bitumen.

The improved adhesion bonding of the aggregate particles is attributed to the increase in viscosity of the NR modified asphalt [56,57].

3.4. Low temperature cracking

Low temperature cracking of asphalt pavement is as a result of thermal shrinkage of the asphalt binder at freezing conditions. This happens when the induced thermal stress at low temperatures surpasses the tensile strength of the mix. Low temperature cracking mostly affects HMA with high stiffness or rigidity. To reduce this type of failure, it is essential for the asphalt binder to have good flexibility of absorbing thermal stress without cracking [46]. Billiter et al., [15] examined the thermal cracking resistance of NR modified bitumen of four different grades of asphalt binder. The study found that NR has positively improve the low temperature performance of all the four asphalt types tested. The improvement is directly proportional to the amount of rubber added, the higher the rubber content the greater the cracking resistance of the binder. Also, smaller size of rubber particles was found to be more resistant to thermal cracking than the larger ones.

3.5. Morphology of NR modified asphalt

For less severe mixing condition, NR in latex form is commonly used as an asphalt modifier compared to the dry form. This is because the latex is ready to be dispersed in liquid asphalt at temperature below 150 °C using low mixing speed (500 rpm) and less mixing time (30 minutes). In principle, NR addition in asphalt tends to physically disperse in not chemically react with asphalt. Thus, the obtained asphalt properties are marginally improved and some properties like penetration and softening point require rather large amount of the NR (up to 10% by weight) to meet the specification. The dispersion and compatibility of NR latex in asphalt using fluorescence microscopy at 400 magnification is shown in Figure 4.

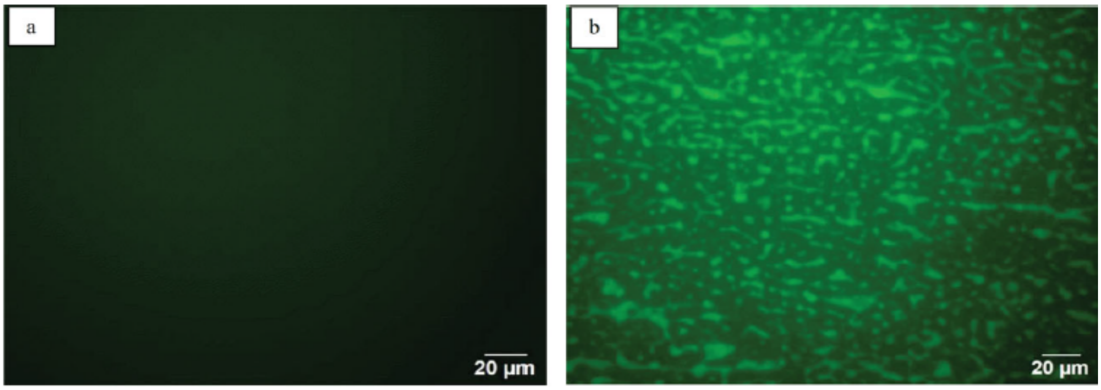


Figure 4. Fluorescent micrographs of NR-modified asphalt (a) unmodified AC 60/70 asphalt, (b) 3.2% NR [12].

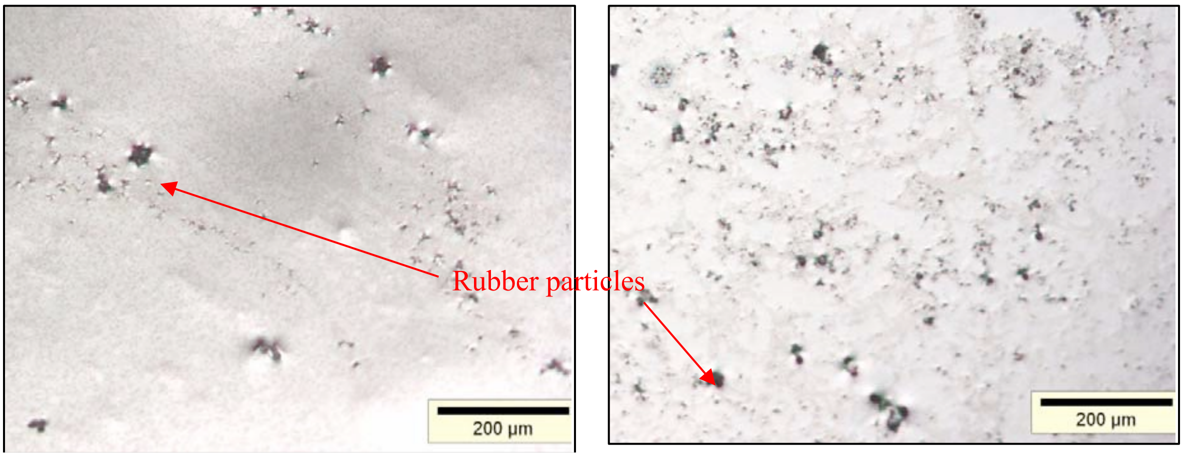
The micrograph above relatively depicts fine NR domains to uniformly disperse medium in the asphalt matrix, indicating a good homogeneity with the domain size of NR of approximately 10 µm. A similar study on the dispersion of NR particles in bitumen using atomic force microscopy (AFM) identified a network of extensive microstructures mixed with bubbles under heat. The AFM images show that the rubber particles size was less than 50 nm and well dispersed in the modified binder [40]. When compared with crumb rubber-modified bitumen the latex particle is considerably smaller in size, smaller size particles improve the stability of asphalt binders considering the balance between Brownian and gravity forces.

3.6. Challenges in NR bitumen modification

The high cost of processing NR modified bitumen is part of the reason why application of NR as bitumen modifier was neglected earlier despite its outstanding properties. The blending of NR with asphalt binder requires high temperature and high shear rate before a homogenous mix of the blend is achieved [58,59]. This consumes a lot energy and sometimes results in degradation of the polymer. Furthermore, the blending process requires special plant mixing or modification of the existing equipment to carry out the task [60,61]. Likewise, the heating effect causes short term ageing of the asphalt binder and generates a lot of greenhouse gases to the environment. However, it should be noted that the cost of periodic maintenance of unmodified HMA pavement will counterbalance the initial high production cost of the NR modified pavement.

Another challenge of modifying bitumen NR is segregation of the polymers in bitumen. Read & Whiteoak [62] pointed out the difficulty of crumb rubber dispersion in bitumen, the ineffective dispersion often results in having a heterogenous blend with the rubber purely acting as an elastic filler. Although NR (in dry form) faced similar homogeneity issue, it shows better reactivity than crumb rubber [63]. Theoretically, NR modified asphalt pavement should have a longer life span than crumb rubber modified asphalt. Though the actual effect needs to be determined through studies and field trials [64]. Generally, pure polymers are thermodynamically incompatible with bitumen, the incompatibility is as a result of the differences in molecular weight, density, polarity, and solubility of the polymer in bitumen [65]. This often results in delamination of the PMB during thermal storage and adversely affect the material during construction [66]. Therefore, it is critical to overcome this drawback if NR is to be used in asphalt modification [36].

Nowadays, solvents like toluene or nano-alumina are used to aid in dispersing the NR in bitumen. Toluene is an aromatic solvent that is widely used in the laboratory for extraction of tar sands and separation of water from bitumen, it also has the ability of dissolving the asphaltene fraction in bitumen [67,68]. Ismail et al., [69] examined the effect of using toluene to increase the dispersion of NR in bitumen. The study immersed 5% NR (by weight of bitumen) in toluene at room temperature for 24 hours, the treated rubber was then blended to 80/100 penetration grade bitumen at 160 °C for one hour at 2000 rpm. Results from rheological tests shows that toluene does not affect the rheological properties of the blend when compared with the conventional high shear blending process. Furthermore, the smaller rubber particle size (Figure 5a) confirms that toluene effectively aids in dispersing the rubber particles more than the conventional high shear mixing process (Figure 5b). The study conclude that toluene improved the compatibility of NR modified binder and does not have any negative effect on the performance of bitumen.



(a) (b)

Figure 5. Fluorescent images of NR modified Bitumen (a) low shear rate with toluene. (b) high shear rate without toluene [69].

3.7. Introducing NR in warm mix asphalt

While there is a great deal of promise that comes along with modification of bitumen with NR; there are concerns about the high energy consumption and greenhouse gas emissions. This is because previous laboratory studies and field trials conducted on the use of NR modified bitumen applied it as a hot mix asphalt. The addition of NR increases the viscosity of the bitumen which in turn requires high temperature during mixing (170 – 200°C), and 150 °C for compaction. This generates a lot of greenhouse gases thereby polluting the environment, it also consumes more fuel during heating which leads to the overall increase in the cost of asphalt production.

A typical example of this is when the Malaysian Rubber board (MRB) and JKR signed a MOU on using cuplump rubber as an additional material in road construction in 2015. Part of the MOU was the construction of three test sections of Cuplump rubber Modified Asphalt (CMA) in Tampin, Negeri Sembilan; Baling, Kedah; and Temerloh, Pahang. After two years of monitoring, results obtained from field and laboratory indicate improvement in resilience, durability, resistance to cracking and rutting over the conventional HMA [70]. Despite its outstanding performance; its application as hot mix asphalt results in increasing the asphalt production cost from RM 29, 900 of conventional HMA to RM 53,600 per kilometre of CMA. The 80% cost increase may be related to the high amount of heating required during mixing and compaction of the CMA. This is because any increase in mixing and compaction temperature of asphalt mixes significantly increases the total energy consumption, which translates to higher asphalt production cost. It is thought that by converting CMA into WMA, the production temperature, carbon emission and cost will be drastically reduced while maintaining the performance of the cuplump rubber modified asphalt mix. This though is in line with the findings by Golchin et al., [71] where the total energy consumption of PMB mixtures is higher than conventional mixtures.

A similar concept of using an elastomer in WMA was conducted by Oner et al. [72] where 50/70 penetration bitumen was modified with SBS, and applied in Sasobit and rediset WMA. The rheological test results show that all SBS modified warm mix asphalt binder has lower temperature susceptibility than the HMA counterpart, also significant increase in rutting performance was observed in all the warm SBS modified binders. Therefore, because cuplump rubber is an elastomer its application in WMA will yield better results than in HMA. Warm-mix asphalt (WMA) represents technologies that allow considerable reduction in mixing and compaction temperatures of asphalt mixes up to 50 °C [73]. Since its inception many agencies around the world have shifted from producing hot mix asphalt to warm mix asphalt. This is achieved by incorporating additives that lowers the bitumen viscosity or expands its volume which in turn helps the aggregates to get completely coated with bitumen at temperatures lower than HMA [74–76]. Some of the benefit of WMA include reduction in fuel consumption, carbon emission, bitumen oxidative ageing, and cracking. Others include longer paving season and hauling distance, early opening to traffic and improved working condition of the workers [77,78] .

Recently, an attempt was made by two independent researchers [79,80] to determine the physical properties NR modified bitumen incorporating Evotherm and Sasobit warm mix additives respectively. The results show that the addition of NR exhibit good dispersion and provides significant increase in softening point and viscosity of the binder while penetration was effectively reduced. The rotational viscosity, mixing and compaction temperatures of the binders rises with increasing NR content. But the addition of warm mix additives slightly reduced the viscosity of the binder, thus suggesting the possibility of using lower mixing and compaction of the asphalt rubber. Surprisingly, both Evotherm and Sasobit modification were found to increase the softening point temperature of the NR modified bitumen. Hence signifying a better rutting resistance of the binder. However, only the mixture evaluation of the modified binders can determine the actual performance of this warm rubberised asphalt.

4. Conclusion

Sustainable roadway construction of today is enhanced through green technology and the best approach is by considering renewable natural resources in binder modification or replacement. From inception; NR modified bitumen was found to enhance the fatigue resistance, temperature and moisture stability of flexible pavement by thickening the bitumen thus increasing its rutting resistance. It also reduces the associated low temperature cracking of the binder through stress absorption. Furthermore, NR provides better cohesion and adhesion of bitumen with aggregate and improve the elastic recovery of asphalt binder. As a result, the road surface becomes more durable and distress free. NR possesses high wet gel strength; it has low cost and has excellent physical properties. Despite the several advantages of NR modified bitumen there are concerns about its higher energy consumption, generation of unwanted pollution and short-term ageing during HMA manufacturing process. In view of that clean asphalt production should involve lowering the manufacturing temperature without compromising the level of mechanical performance through WMA. Some of the advantages of applying NR in WMA include lower mixing and compaction temperatures, 30 - 50% reduction in fuel consumption and significant reduction in the amount of CO_2, CO, NO_x and SO_2 released to the environment. The energy savings translates to lower construction cost and a more environmentally-friendly road construction setting for the workers.

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