

Cup Lump Natural Rubber

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Cup lump is obtained when fresh latex is extracted by tapping into a long cut made in a rubber tree and letting the drips pour into a plastic cup.

energy

sustainability

natural rubber

bitumen

additives

rubberised bitumen

1. Introduction

Cup lump is obtained when fresh latex is extracted by tapping into a long cut made in a rubber tree and letting the drips pour into a plastic cup. The substance is then mixed with formic acid to coagulate the latex. The process continues for several days until the required amount is reached ^[1]. Physically, CLNR consists of water, rubber and non-rubber material. The moisture content of CLNR often varies locally because of surface evaporation, which creates a moisture gradient that encourages diffusion toward the drier surface. According to Pamornnak et al. ^[1], the volume of CLNR generally ranges between 500 and 1500 cm³ depending on the cup used for collection. Normally, CLNR is around 76 mm high, with a diameter of 38 mm at the bottom and 101 mm at the top. It has a low water content and chemical properties identical to NRL, which have prompted researchers to explore its use in bitumen ^[2].

S. Abdulrahman et al. ^[3] mentioned that CLNR content significantly affects the storage stability and viscosity of the cup-lump-modified bitumen (CLMB). The rise in bitumen stiffness is proportional to the increase in mixing and compaction temperatures. In addition, the use of CLNR with additives produces excellent rutting resistance, improved rheological properties, better viscoelastic response at higher temperature, improved tensile property, moisture resistance and enhanced conventional properties of the base bitumen, such as penetration, softening point and temperature susceptibility ^{[4][5][6][7]}. An analysis of physical and rheological properties showed that the presence of elastic NR within the bitumen network increases the softening, viscosity and stiffness, and it reduces penetration and temperature susceptibility, consequently enhancing the rutting property ^{[5][6][7]}. Othman et al. ^[8] conducted an on-site evaluation and found that cup-lump-modified asphalt (CMA) pavement performed better in surface conditions with a high roughness value, structural condition, skid resistance and dynamic creep.

The physical properties of the rubber have a significant influence on the composition and viscosity of bitumen; as a result, the physicomechanical properties of bitumen pavement are influenced by the rubber polymer, which is primarily regulated by rubber–bitumen interactions. Jeong et al. ^[9] mentioned that these interactions vary based on the bitumen composition and the rubber surface treatments, and they are sensitive to environmental variables.

Additives help improve the performance of CLNR in bitumen modification. Choosing an appropriate additive varies from one area to another depending on the geographic location and resources in each country. Pavement professionals should not choose additives based solely on how well bitumen functions; instead, they should consider other aspects, such as economic concerns, the manufacture of modifiers and environmental compatibility [10]. Previous studies indicated that one modifier alone cannot enhance all the requisite pavement functional properties. Bitumen should be modified with several additives to improve diverse characteristics because of the complex interactions of modified bitumen.

2. Treatment of Cup Lump

A homogeneous mixture is crucial to ensure a uniform composition that cannot be separated physically through any separation process. Generally, the components of a homogeneous mixture retain their own properties. Polymer-bitumen compatibility is crucial to achieve a homogeneous mixture by including well-dispersed polymer into bitumen formulations. CLNR is initially solid; thus, a pre-treatment technique was suggested to promote the dispersion of CLNR into bitumen. CLNR should be pre-treated with a chemical solvent to soften it before being used as a bitumen modifier. In practice, the solvent evaporation behaviour makes it less prominent in modified bitumen, which keeps the bitumen characteristics from being significantly disrupted. Studies have examined the interaction of rubber with aromatic hydrocarbons as a chemical solvent for surface treatment, and rubber swelling to alter its shape as a sticky material [11][12]. According to previous studies, a combination of evaporation-prone chemical solvents such as toluene and xylene can be used as a rubber solvent. According to the Occupational Safety and Health Administration, toluene is a clear, colourless liquid that turns into a vapour when exposed to air at room temperature and has a distinct odour. Xylene (C_8H_{10}) is a colourless, flammable liquid with a sweet odour. Toluene and xylene may be used in bitumen. However, few studies have evaluated how their use affects bitumen modification.

Referring to Azahar et al. [13], various rubber-to-toluene ratios at 1:1, 1:1.5, 1:2, 1:2.5 and 1:3 with 20 g of cup lump were placed in five separate steel containers with lids, to identify the optimum ratio. The samples were kept at room temperature in the lab, and CLNR's swelling behaviour over time was observed while the rubber-to-toluene ratios and the treatment time were analysed. Steel containers with identical dimensions were employed to verify that the rate of evaporation was consistent. For the first day of treatment, the toluene loss was measured hourly to track CLNR's physical change. The record was continued the next day until the toluene had entirely absorbed and evaporated. The 1:2 rubber-to-toluene ratio was identified as the optimum ratio at the end of the experiment. This optimum ratio corresponds to the findings of Abdul Ghafar et al. [14]. CLNR with a size of 10 mm was treated with a toluene-to-CLNR ratio of 2:1 for 24 h before being mixed with base bitumen. The same ratio was used by Mohd Azahar et al. [15]. Next, 5%, 10% and 15% rubber crumb were prepared with toluene for 24 h with a 1:2 rubber-to-toluene ratio. The blending procedure consisted of two steps. First, the treated rubber was shredded. Then, it was combined with bitumen. The treated rubber was initially placed in the centre of the supporting column, which consists of a shaft attached to the motor at one end and the head at the other. The treated rubber was rotated for 2 min at 2000 rpm with the intention of ripping the rubber surface. This method might slow down the ageing of

bitumen and ensures that toluene is discharged by evaporation because of the shorter duration needed to integrate the two ingredients.

In the application of xylene, Hazoor Ansari et al. [2] treated 50 g of dried CLNR with different CLNR/xylene ratios of 1:1, 1:2.1:3 and 1:4. All containers had equal sizes and dimensions to prevent any variation in the outcomes. All containers were stored at room temperature in a lab on a flat, level surface for 48 h. The optimum ratio was 1:3. CLNR was gradually introduced into the bitumen in increments at a shearing speed of 500 rpm to increase the xylene evaporation from CLNR. This procedure can shorten the blending time, enhance rubber absorption and eventually slow down bitumen ageing. The swelling and deterioration of the rubber often cause rubber–bitumen interactions at high temperatures. Only the broken down and disentangled CLNR chains interacted with the heated bitumen in pre-treated CLNR because the solvent already regulated the swelling. The low-molecular-weight portion of bitumen (maltenes) is disseminated and absorbed by the network of polymers when CLNR interacts with warmed bitumen at a high shearing speed and temperature; this process encourages additional xylene evaporation.

3. Type of Additives and Their Methods of Application

The use of additives in CLNR mainly aims to increase the homogeneity of the bitumen, thus enhancing the properties of the mixture for better pavement construction. A high CLNR content corresponds to severe mixing conditions and thus a higher viscosity of the base bitumen. This condition requires higher mixing and compaction temperatures, resulting in poor flow characteristics for the modified bitumen. Furthermore, this condition produces a large amount of polluting fumes and oxidative ageing, which will increase the overall cost of bitumen production [3][4]. Moreover, polymer has significant drawbacks, such as degradation, oxidation and susceptibility to free radicals in rubber, which accelerate bitumen ageing [16]. Therefore, for a clean manufacture of rubberised bituminous mix, the manufacturing temperature should be reduced without sacrificing mechanical performance. For this reason, a wide range of additives that can increase distinct characteristics of bitumen and work in diverse ways have been utilised in rubberised bitumen modification, such as PPA, Evotherm, MPP, TMQ and sulphur, which were identified as co-modifiers [3][5][6][11][17][18][19].

3.1. Polyphosphoric Acid

PPA is a reactive agent made of liquid mineral polymer with a series of phosphoric acid oligomers that has gained attraction due to its relatively enhanced bitumen performance at a low cost [20][21][22]. The addition of PPA shows good compatibility with the bitumen and can be used in conjunction with other bitumen polymer modifiers such as styrene–butadiene–styrene (SBS) and styrene–butadiene rubber (SBR), ground tire rubber (GTR), desulphurised rubber-modified asphalt (DRMA), crumb rubber (CR), and latex-modified bitumen [20][23][24][25][26][27]. Unlike SBS, economical modifiers such as PPA can improve the characteristics of bitumen. Numerous functional groups in bitumen can react with PPA [27]. It disperses the asphaltenes and makes improved interactions with rubber possible by allowing for greater dispersion of asphaltenes in the maltenes phase [28]. Using a modest quantity of PPA to

create a PPA-modified bitumen will give it rheological qualities comparable with those of an SBS-modified bitumen [23].

Hazoor Ansari et al. [2] examined how CLNR and PPA affect bitumen properties, including its shape, rheological features, temperature susceptibility and storage stability. Modified bitumen was obtained by adding four percentages of CLNR (3%, 6%, 9% and 12%) and PPA at 0.5% by weight of bitumen and mixing them for 30 min. After PPA was blended with base bitumen at concentrations of 0.5%, 1%, 1.5% and 2%, the optimal value was determined to be 0.5% by trial and error. After 90 min of mixing, the mixtures were held at 180 °C for an additional three hours. This method was used to ensure that there was no trapped air and to further disseminate the rubber chains throughout the bitumen for a uniform modified bitumen. The greater PPA concentration with CLNR was impractical because it also causes the base bitumen to stiffen [29]. Saowapark et al. [30] mentioned that the modified bitumen with 3.2 wt.% of NRL and 2 wt.% of PPA is a recommended formula that is suited for the road conditions in Thailand with a fixed shearing time of 30 min. In addition, Qian et al. (2019) [31] determined 1 wt.% as the ideal dosage of PPA in CR-modified bitumen at a shearing speed of 600 rpm for 30 min. This result was similar to that obtained by Yadollahi and Sabbagh Mollahosseini [28]. Bitumen was mixed with 1% PPA at 160 °C for 60 min while PPA was gradually heated to 175 °C. Many bubbles form during PPA blending, indicating that bitumen and PPA undergo a chemical reaction with some gas. Therefore, mixing should not cease until all bubbles have vanished. Then, 1% PPA was added to bitumen to catalyse the formation of low- and high-sulphur bitumen [32].

3.2. Evotherm

Warm-mix asphalt (WMA) technology has grown in popularity in recent years because it can drastically lower the working temperature of the bitumen mixture while ensuring its performance and minimising pollutant gas emissions during the mixing and paving stages, resulting in improved on-site construction conditions [33][34][35]. Other advantages of WMA technology include lesser fuel consumption, a faster construction period and transportation time opening and less bitumen thermal ageing [17]. Evotherm warm-mix additive is a chemical package that comprises workability accelerators, adhesion promoters and emulsifiers [20]. Evotherm is a brown, oily liquid or chemical additive that is partially water soluble and has a fishy amine odour. It contains amine agents that increase bitumen workability during the mixing and compacting process. The product allows bitumen mixes to be mixed and compacted at lower temperatures without compromising quality.

Suleiman Abdulrahman et al. [36] conducted a study using CLNR and with 0.3%, 0.4%, 0.5%, 0.6% and 0.75% Evotherm-modified bitumen at 160 °C for 5 min to produce warm cup-lump-modified bitumen (WCMB). The optimum WCMB was identified and the ideal mixing and compaction temperatures were established through physical and rheological tests, which were conducted to evaluate the impact of this alteration by using aggregate coating and compatibility tests. A similar study was conducted by S. Abdulrahman et al. [3] but with different shearing temperature, speed and time at 170 °C and 4000 rpm for 2 h, respectively. Then, 0.5% Evotherm was selected as the optimum dosage of the warm-mix additive. Furthermore, the CR-modified bitumen in its terminal blend was studied with 0.3% dosage of Evotherm by weight of bitumen [21]. H. Yu et al. [22] used Evotherm to investigate the rheological properties of warm rubberised bitumen prepared by different mixing procedures at 160

°C for 1 h. H. Yu et al. [37] used different shearing times. Their blending process was conducted for 10 min at 160 °C with a shearing speed of 800 rpm.

3.3. Mangosteen Powder

Rahmah et al. [38] evaluated the effect of 2–6 g of MPP on the ageing properties of 10 g CLNR. MPP has different bioactive compounds such as phenolic acid and flavonoids, which have biological and medicinal properties. Notably, antioxidant capabilities are due to phenolic natural xanthenes such as α -mangostin and γ -mangostin [39]. MPP is applied mainly because it can replace synthetic hindered phenols or amine and enhance the ageing properties of the rubber modifier [38]. MPP also helps prevent oxidative degradation [40].

3.4. Trimethyl-Quinoline

TMQ (2,2,4-trimethyl-1,2-H-dihydroquinoline) is an excellent antioxidant that is relatively low cost and has various uses in rubber manufacturing processes. It reacts with oxides or broken polymer chain ends generated by reaction with oxygen [41]. Its product is a combination of oligomers such as dimers, trimers and tetramers. The content of oligomers, particularly dimers, determines the quality of TMQ [42]. As a result, they prevent oxidative degradation from spreading, thus preserving the elastomer physical properties. Used polymers are usually disposed of in the environment, and can be recycled in some circumstances. Thus, additive optimisation is essential for the distinctive qualities of rubber to function and for the release of various additives into the environment to be controlled [43].

3.5. Sulphur

The application of sulphur on CLNR has not yet been reported, but its significant contribution to the enhancement of properties has been documented by previous researchers. The low cost and availability of sulphur as a by-product of the oil and gas sectors may explain the spike in interest in using it as an additive or partial alternative for bitumen [44]. Since the 1940s, sulphur has been found to affect and improve the characteristics of bitumen pavements [45]. When sulphur is used as a bitumen extender, it decreases the amount of bitumen needed and greenhouse gas emissions while also changing the rheological properties of bitumen. As a result, it minimises the use of bitumen, a naturally occurring substance on the verge of depletion, and makes the mix more cost-effective because sulphur costs three to four times less than bitumen [33]. The influence of sulphur on bitumen varies depending on the bitumen's origin, the types of sulphur used, the sulphur concentration and the blending temperature [46][47]. When the price of bitumen returned to normal, sulphur in pavement engineering became less appealing, and safety and environmental issues have been raised about processing sulphur-extended bitumen. Sulphur can accelerate the cross-linking action between the polymer and the components in the bitumen and establish a strong chemical connection, which manifests as a microscopic impact of a more uniform rubber polymer phase distribution [47].

According to Saowapark et al. [48], 0.3 wt.% of sulphur sheared for 10 min with 3.2 wt.% of NRL was selected as the optimum concentration for the road conditions in Thailand. A similar dosage was used by S. Wang et al. (2021) [49], showing that sufficient sulphur concentration helps ensure that the network structure of the CR-modified

asphalt (CRMA) is homogeneous. However, too much sulphur will prevent the desulphurisation and deterioration of the CR in the bitumen, which would harm the CRMA's low-temperature performance. The blending process was performed under a shearing time of 120 min at 180 °C and is similar to that performed by S. Wang, Huang, Liu, Lin et al. [50]. In the application of SBS, 1% sulphur was added to 2% of SBS with stirring for 2, 4 and 8 h. Gupta et al. [51] also used sulphur as a cross-linker in the fabrication of new experimental bitumen using bitumen (70/100) and high-biaryl content styrene-butadiene copolymer. Hung et al. [52] mixed rubber-modified bitumen with 3 wt.% sulphur at 135 °C for 5 min. The same shearing condition was implemented by Mousavi and Fini (2021) [44], in which 10% sulphur was blended with rubberised bitumen by weight. The same concentration of sulphur (10% by base bitumen weight) was blended at a shear speed of 3000 rpm at 180 °C for 30 min, with the bitumen produced having a high sulphur content [46][47]. Zhou et al. [53] added 10% sulphur to bio-modified rubberised (BMR) bitumen at a temperature of 155 ± 5 °C for 30 min using a mixer at 1000 rpm. Das and Panda [54] added 2% of sulphur to bitumen at various temperatures ranging from 100 to 160 °C, with a 10 °C increment for various mixing times of 15, 30, 45, 60 and 75 min.

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