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Study of storage stable bituminous mixture made with crumb rubber modified bitumen

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ABSTRACT

The surface activation of crumb rubber particles was carried for effective dispersion of it into bituminous matrix. The resulted activated CRMB was examined in terms of its consistency and conventional properties as per existing specifications. The storage stability studies are also taken into consideration, in which softening point values were taken from top and bottom portion of the specimen after 48 hours storage at 163⁰C. The difference in softening point of top and bottom portion of the activated CRMB is found only 2.5⁰C, showing that the rubber particles are homogeneously mix with bitumen. The increased value of softening point and a decrease in penetration value of activated CRMB also exhibited the improvement in viscosity at working temperature and stiffness of the bitumen binder, and it also meets the requirement standard specification.

Keywords: CRMB, storage stability, waste tyre rubber, asphalt mix

1. INTRODUCTION

It is very difficult to maintain the fluidity and workability of bituminous mixture (asphalt mixture) at manufacturing plant as well as at working site because it is affected by many factors such as shape, sieve size and texture of aggregate, grade of bitumen binder, binder content, mixing and temperature. Workability of asphalt mixture can be related to the compatibility between bitumen binder and additives used to enhance the physical and rheological properties of asphalt mixture. Therefore to achieve the desired physical and mechanical properties of bitumen binders to be used in flexible pavement, some additives or modifiers/polymers are added to it. In polymer modification of bitumen, the polymer or modifier is incorporated in bitumen by mechanical blending or chemical reaction[1]. Keeping in mind the cost of flexible pavement, crumb rubber is best modifier[2-8].

A report, The Future of Global Tires to 2022 estimates the production of tyres 2.7 billons units in 2022 with growth of 3.4% annually[9] and around 1billons of waste tyres are generated in each year out of which only 100 million tyres are recycled industrially[10]. This represents 2-3% of total waste collected[11]. The total tyre production in India was around 169 million units in the year 2021[1].

The tyre is much more complex and technically safe innovative product as seen in figure 2 and innovation in tyres is still on-going. Passenger tyres are usually constructed with five components: (a) Tread is made up of rubber compounds and this part makes contact with the road, (b) Side wall of tyre covers between the tread and bead and also made up of rubber compounds, (c) Body ply is made up of synthetic fibres like polyester and rayon and it forms the skeleton of tyre, (d) Belt is made up of stiff steel wire and give stiffness to the tread and protect the carcass and (e) Bead is designed to grip the tyre to wheel base. Bead is made up of bundle of fine steel wire[13]. This shows that tyre is made up three types of materials, elastomeric compound, fabric and steel[14].

In wet process, the crumb rubber is mixed with asphalt binder (mix of bitumen, aggregate and other additives), and known as rubberized asphalt binder, which deliver enhanced performance and also extended service life to the pavement, resistance to fatigue, thermal stability, reduced noise[31-35]but segregation of crumb rubber from asphalt binder limits its application[36, 37], due to high setting rate of crumb rubber. In dry process, crumb rubber is added to the aggregate and then mixed with bitumen. In this case, the interaction of crumb rubber with bitumen binder is very less. The crumb rubber consists of vulcanised rubber, causing inadequate dispersion and incompatibility in asphalt binder. The phase separation in asphalt binder also caused by density difference of crumb rubber (1.13 g/cm^3) and bitumen (1.02 g/cm^3), and swelling of crumb rubber particle, as they absorbs maltene portion of bitumen[38] creating two phase within bitumen binder; polymer-rich phase and asphaltene-rich phase. The dynamic asymmetry between two phases directly gives rises to phase separation process. The difference in microstructure, physio-chemistry such as glass transition temperature, polarity, molecular weight and solubility for polymer and bitumen also contribute to the phase separation[39].

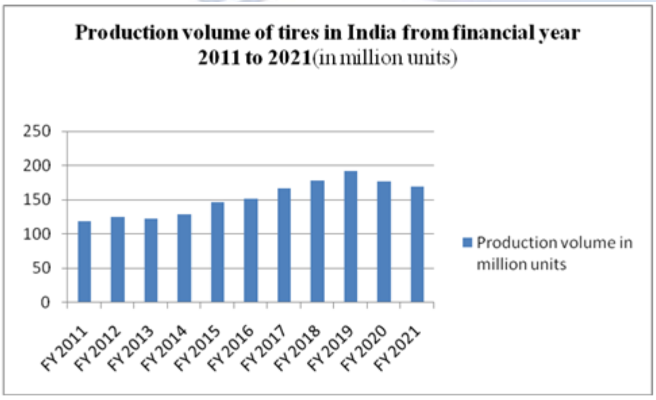

Fig.1. Production volume of tyres in India from financial year 2011 to 2021[12]

The chemical and physical characteristics of the tyre such as resistance to UV rays, heat, humidity, acids, alkalis, some solvents, and non-biodegradability, shape and elastcity are same worldwide. However, many of the properties which are advantageous on road life are disadvantageous, when they are discarded and can create many problems like collection, storage and disposal[15]. Disposal of used tyre is the biggest threat to waste management in 21st century, because their non-biodegradability, flammability turns them into a main source of fumes, habitat for rodents/insects, and also their toxic chemicals pollutes soil and water[16,18, 19, 20]. However, discarded tyres may be disposed by many ways like landfilling, energy, stockpiles, etc, but the best method for their management is recycling to produce crumb rubber. Over the last 25 years, many industries have invested in modern technologies to recycle the old tyres to produce valuable rubber an after removing other components such as steels, fibres and other ingredients from tyres[21-25]. Therefore, by using this rubber as additives in bitumen binder, there is not only the protection of environment, but we can stop the economic loss by using recycled rubber in place of virgin to enhance the performance of pavements[26, 27]. Modification of bitumen with crumb rubber for using in pavement and roofing applications can be achieved by three main processes; wet, dry and terminal process[28-30].

Fig. 2. Tyre structure[13]

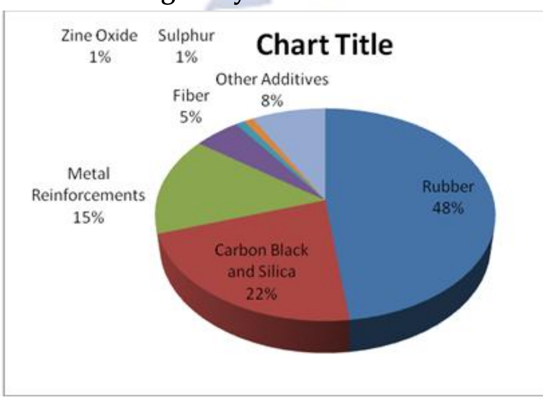

Figure3. A Typical composition of a vehicle tyre[16, 17].

The compatibility between bitumen and polymer additives also depends on the solubility parameters[40]. The extent of improvement in the performance asphalt binder depends on the types and amount of rubber in crumb particles, size of crumb particle, carbon black content, and ratio of bitumen and rubber, source of bitumen and processing conditions like temperature, blending time, and speed of blender[41, 42]. During the blending of bitumen and crumb rubber, vulcanized rubber undergoes limited chemical changes i.e, it is only physical mixing rather than chemical interaction between bitumen constituents and crumb rubber, because it is sulphur cross-link rubber, resulted the sedimentation of crumb at the bottom of test tube during storage stability test[43]. Because crumb rubber is thermoset in nature, the incompatibility problem occurs by the existence of low interfacial force between the crumb rubber particle and bitumen,[44] which affects rheological properties of bitumen binder. If crumb rubber is not partially devulcanized, it acts only as flexible filler in crumb rubber modified bitumen (CRMB). Therefore to make it an effective modifier for CRMB, it is necessary to cleavage the C-S and S-S bonds of crumb rubber.

Since storage stability is the main parameter to access the performance of CRMB, understanding the mechanism and compositional factors is thus of great concern for both production optimization and quality control. Other then process optimization of CRMB, the improvement in phase separation of CRMB may be achieved either by using some chemical compounds to form bonds between different components of bitumen and modifiers forming a polymer network structure[45]or by activating the surface of crumb rubber particles by microwave, ultrasonic, thermochemical, thermomechanical, biological and plasma methods[46, 47]. S. Kocevski et al reported that Furaldehyde or acrylic acid was also used to activate the surface of crumb rubber[48].

In this study both the ordinary crumb rubber and partially devulcanized crumb rubber along with additives was used to modify the bitumen binder for paving applications. The main objective of using activated crumb rubber as modifier is to obtain a homogenous binder in which crumb rubber particle are dispersed uniformly throughout the bitumen phase. The resultant binder would provide improved elasticity, improved temperature susceptibility, strength and desired fluidity compared with ordinary crumb rubber modified bitumen.

2. EXPERIMENTAL

Materials

In this investigation, the penetration grade bitumen (penetration, 97 dmm; softening point, 46^{0}C; elastic recovery, less than 7%) was taken from a local market produced in nearby Indian refinery.

The crumb rubber produced by an ambient process was purchased from a local recycling plant. The physical and chemical properties are given in table 1. The cross-link density of obtained crumb rubber was 6.4 mol/cm^3. The gilsonite, high molecular weight and polynuclear hydrocarbon, available in local market, was taken to as modifier (softening point 155^{0}C; penetration 1.8 dmm; solubility in CS_2 about 86wt %). Gilsonite, being a natural asphalt, it readily dispersed into bitumen phase to form a stable binder.

Table1. Physico-chemical properties of crumb rubber[49]

S.N.	Properties	Values
1	Density	1.13 g/cm^3
2	Moisture content	0.61%
3	Carbon black content	28%
4	Rubber hydrocarbon content	52%
5	Acetone extract	10.8%
6	Ash content	7.3%

Methods

The partial devulcanization of crumb rubber was carried out through thermo-mechanical methods in an open two roll mixing mill for 2-3 hours in the presence of petroleum tar, at the temperature range of 220-230^{0}C, resulting 17-18% devulcanization of crumb rubber. This crumb rubber is mixed with elemental sulphur and then with gilsonite to obtained dry crumb rubber concentrate.

To prepare CRMB binder, this treated crumb rubber was gradually mixed with the bitumen, which is preheated at 160^{0}C in a closed steel vessel fitted with higher shear mixer. The concentration of crumb rubber in CRMB was optimized about 10 wt % keeping in view of its workability and available specifications[50].

Test Methods

Softening point test

The softening point is the temperature at which the bitumen attains a particular degree of softening. It is measured by a Ring and Ball apparatus as per ASTM D 36-09. In this test, bitumen is heated approximately above its softening point, and then melted bitumen was poured into the rings placed on the glass plate, coated with a mixture of glycerine and dextrin. It prevents the bitumen from sticking to the plate. Now allow the rings to cool at room temperature for about 30 minutes in air. After that the rings were placed in apparatus and heated in glycerine bath at a constant rate of 5^0C/ min. The temperature at which bitumen filled in the rings soften and sag downward by the weight of standard steel ball, is noted as softening point.

Penetration test

It is measured by using the apparatus known as Penetratometer, to know the value of hardness or softness of bitumen. This test is performed according to the ASTM D 5-06, in which 100 gram of load is applied on the penetration probe for 5 second at 25^0C and reported the penetration of needle in bitumen in $1/10^{th}$ of millimetres.

Elastic recovery Test

The elastic recovery of base bitumen and CRMB is determined the apparatus known as Ductilometer, according to IS: 1208 – 1978[58]. For elastic recovery test, the test specimen is elongated to a deformation 10 cm at a speed of 5± 0.25 cm/minute at 15^0C then immediately cut the test specimen into two halves at the midpoint. Now after keeping the test specimen in the water for an hour, length of the recombined specimen is recorded by moving an elongated cut half of the specimen back into position just touching the fixed half the test specimen.

Cigar tube test

This test is widely carried out to know the degree of separation between the polymer phase and bitumen during hot storage in laboratory according to ASTM D 7173, and this can be evaluated by comparing ring and ball softening point of the top and bottom portion of the conditioned sample[52]. For this test, firstly the 50 gm of specimen is poured into aluminium tube, after heating it up to 163^0C. The sealed tubes are placed vertically inside an oven at 163^0C for 48 hours. Then these tubes are cooled at -18^0C in a freezer for 4 hours, taking care to keep the tubes in vertical position all times. After that the tubes are cut into three equal pieces. The sample from both the top and bottom portion of test tube are evaluated for phase separation studies of polymer or rubber modified bitumen such as softening point test, dynamic shear rheometer, morphology and microstructure.

Results and Discussion

The gradation of activated and ordinary crumb rubber particles is given in figure 4, which shows that the activated crumb rubber particles were fine as compared to ordinary crumb rubber particles and uniformly distributed. The retention of activated crumb rubber particle is 74.84% on a 150 µm sieve as compared to 97.5 % retention of ordinary crumb rubber on the same sieve and the average particle size of activated crumb rubber and ordinary crumb rubber is 255 µm and 375 µm respectively. This shows that crumb rubber have altered their surface and thermosetting character due to devulcanization. The devulvanized crumb could provide an elastomeric and storage stable CRMB, which is proved by storage stability test data given in table 2. A bituminous system is called to be stable, if the difference in softening point of the bottom and top portion is less than 3^0C[53], after a long storage at elevated temperature.

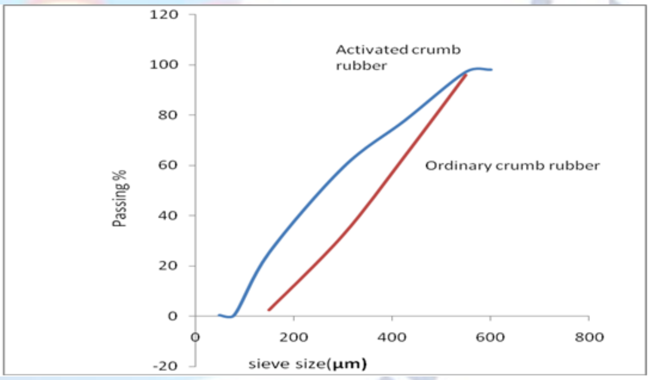

Fig.4. Gradation of activated and ordinary crumb rubber particles (passing % Vs Sieve size)

It is concluded from the table 2 that the ordinary CRMB exhibited the difference in softening point of the top and bottom sections 28^0C as compared to 5^0C for base bitumen, suggesting a substantial phase separation between the crumb rubber and bitumen. This difference of temperature shows that the crumb rubber particles are not dissolved in bitumen matrix and poor compatibility between them has led to phase separation, due to migration of crumb particles to top section.

Contrary to this, activated CRMB, displayed the difference in softening point of the top and bottom sections only 2.5^{0}C as compared to 5^{0}C and 28^{0}C for base bitumen and ordinary CRMB respectively. This indicates that surface activation of crumb rubber particles contributes in the improvement of storage stability of activated CRMB. The slight difference in softening point of the top and bottom sections in case of base bitumen may be due phase separation of asphaltene and maltene phases of bitumen.

Table 2. Storage stability test of base bitumen and CRMBs

Specimens	Softening point (^{0}C)		Difference (^{0}C)
	Top	Bottom	
Base Bitumen	55	50	5
Ordinary CRMB	90	62	28
Activated CRMB	66	63.5	2.5

The conventional properties of base bitumen and CRMBs are given in table 3. Increase in softening point and decrease in penetration values are observed in for the activated CRMB in contrast to the above values for ordinary CRMB, exhibiting the improvement in properties of bitumen binder. The decrease value of penetration shows that the binder becomes harder and stiffer by addition of crumb rubber into it. Similarly the increase in softening point values of activated CRMB is due to improvement in the viscosity of it. The increased value of elastic recovery also confirms rubberized network in bituminous system.

Table 3. Conventional properties of ordinary and activated CRMB as per IS: 15462 - 2004[50]

Property	Ordinary CRMB	Activated CRMB	Specified Value IS: 15462 - 2004
Softening point (^{0}C)	62	65	60
Penetration (dmm)	56	39	Less than 50
Elastic recovery (%) at 15^{0}C	51	57	50 (min.)

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3. CONCLUSIONS

The result indicates that crumb particles became finer after their surface activation and led to uniformly dispersion in bitumen. This is why, the activated CRMB displays good storage stability. It was observed that the use of activated crumb rubber particles provide superior consistency, rheological and conventional properties to the base bitumen than the use of ordinary crumb rubber particles in bitumen. It is suggested that incorporation of activated crumb rubber in bitumen can be a better option for waterproofing and paving applications. It can be a best alternative to virgin rubber/polymer for modification of bitumen, and also provide a handsome saving due low price of crumb rubber as compared to virgin polymer.

Conflict of interest statement

Authors declare that they do not have any conflict of interest.

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