CHAPTER 2
LITERATURE REVIEW

The literature survey was carried out for the understanding of the research work done in past few years on the bitumen modification for improvement of overall performance of bitumen for road pavement and the methods used for bitumen modification. In the first section, we have emphasized over the basic understanding for bitumen modification by crumb rubber. In the second section, the complete literature survey was conducted for crumb rubber modification using various types of additive to improve storage stability with increased rheological properties and with their benefits.

2.1 CRUMB RUBBER MODIFIED BITUMEN (CRMB)

Bitumen is a highly viscous material which has good mechanical and adhesive nature; it is preferred in road construction. However, the strength of bitumen is highly affected by environmental factors, which causes stripping, fatigue, rutting in the bituminous pavement, thermal cracking. Therefore, to meliorate the properties of bitumen, various polymers and additives were used in bitumen modifications, i.e. antistripping chemicals, reactive polymer, thermoplastic & thermoplastic polymer, scrap crumb rubber. A polymeric modification of bitumen has been increasing in bituminous pavements for road construction.

Economic growth, expanding population and accompanying structural changes strongly lead to increase transportation sector. With increased automobile industrial world, disposal of waste tires is a major concern throughout the world. Recycling and reusing of waste used tires are still challenging and has been considered as a serious threat to green environment and human life. Crumb rubber is most commonly used polymer to modify bitumen using SBS.[12] According to a report published in 2004, with rapid increase in automobile industry, about 112 million tons of scrap tires were produced.[13-14] Crumb rubber is vulcanized rubber, a kind of inert polymer materials produced from used tires used for bitumen modification. The incorporation of scrap tires rubber (crumb rubber) in bitumen pavement mixture by two ways.[15] The mechanical performance of crumb rubber modified bitumen by wet process has more permissiveness to higher deflections with a higher fatigue resistance compared to conventional bitumen.[16-17]

2.2 WET PROCESS

In wet process of CRMB, an interaction between bitumen and CR occured at high temperatures to form a viscous liquid. Several parameters like blending time, blending temperature, type of mechanical mixing of CRMB are responsible for the interaction process and also the size, type and specific area of CR and type of binder.[18]

Farina et al., (2017)[19] reported that crumb rubber modified bitumen produced by wet technology could be an alternate and safe way of waste tyre disposal. They have studied seven different asphalt mixtures including 30% and 60% reclaimed asphalt pavement (by the weight of natural aggregate) were modified by 0, 10%, and 20% crumb rubber powder with high percentage of organic additives.

Feng Zhang et al. (2015)[14] studied the structural properties of modified crumb rubber bitumen using SBS and sulfur. They have shown that CRMB have improved tenacity with SBS addition and the storage stability of crumb rubber or SBS-modified bitumen. Morphological study by FTIR and ^{1}H NMR showed the distribution of functional groups of modified binders.

Peilong Li et al. (2017)[20] showed that analysed the swelling and degradation behaviour of crumb rubber modified bitumen at 20% loading at 195 ◦C with a processing time is less than 1.5 h. They conducted infrared spectra (FTIR) and differential scanning calorimetry (DSC) of crumb rubber and reported that swelling of molecular chains of rubber resulted to disentangle and released some smaller molecule materials, which could be easily dissolved in bitumen.

Nejad et al. (2012)[21] investigated the propertied of improved bitumen using classic and SHRP testing. The result showed that addition of crumb rubber has come down the penetration, ductility, and stepped up the SP, ER.

Baha Vural Kök et al. (2011)[22] carried out a research to compare the crumb rubber modified bitumen and hot bitumen mix using rotational viscometer (RV), DSR with conventional tests. The result showed that to accomplish the same performance, like SBS-modification, the high amount of crumb rubber must be used than SBS. They found that 8 wt. % crumb rubber modification was the most suitable content than the 2%, 3%, or 4% SBS modifications. According to stiffness modulus the 8% crumb rubber mixture demonstrated similar performance as for 4% SBS modified mixture with a 50% higher stiffness modulus as compare to base mixture.

Xiao et al. (2009)[23] reported that the incorporation of CR in bitumen was helpful to increase the voids in mineral aggregate in Superpave mix design with improved rutting resistance of bituminous mixtures irrespective of type and size of rubber.

Wong et al. (2007)[24] studied the effect of CR modifiers on high temperature susceptibility of three gradations (AC-10, AC-20 and PA). The course mixture was prepared using 10% crumb rubber modifier in wet process with three sizes, i.e. 0.15 mm, 0.30 mm and 0.60 mm. The result showed that crumb rubber modifier addition contributed to improved performance at high temperatures. And among three 0.15 mm crumb rubber size marched the best effect on the dense-graded mixture (AC-10 and AC-20), while modified CRMB with 0.60 mm CRM presented the outperformed on the bitumen.

Ling Pang et al. (2014)[25] used layer double hydroxides (LDHs) into the Crumb Rubber Modified Bitumen (CRMB) to increase its aging resistance. They have reported that the softening point of modified asphalt was increased and G* ratios was decreased significantly with increased ductility and the penetration due to the introduction of LDHs. Wu et al. (2012)[26] reported the enhanced aging property of modified asphalt after addition of layer double hydroxides.

Paravita Sri Wulandari et al. (2017)[27] used crumb rubber as a modifier for bituminous concrete mixture preparation. They have used two different crumb rubber contents (1% and 2% by weight of bituminous mixture) with two different crumb rubber sizes (0.42 mm and 0.177 mm) and conducted the Marshall test to determine the durability of asphalt mixture. They found that incorporation of crumb increased the strength and quality of asphalt mixture.

Ana MaríaRodríguez-Alloza et al. (2014)[28] studied the temperature properties of modified bitumen with warm mix additives using crumb rubber (20% by modified binder weight). They have also mentioned that incorporation organic additives reduced the viscosity of the modified binder, which allows the reduction in temperature for high viscosity of 2800 cP, the result showed that as 2% or 4% of an additive could reduce the temperature by 10 or 30 °C, respectively, with Sasobit, 7 or 19 °C with Asphaltan A, 4 or 21 °C with Asphaltan B and 2 or 10 °C with Licomont.

Osman Nuri Çelik et al. (2008)[29] tested the workability and compatibility of bituminous hot mixtures prepared by adding the waste tire rubber particles in bitumen with different sizes and varied concentration. They reported that type of binder and viscosity of modified bitumen have great effect on the compatibility and modification with crumb rubber it become less compactable when compared with neat bitumen.

Al-Mehthel (2015)[30] utilized elemental sulfur, and crumb rubber for the purpose of bitumen modification. In this process, crumb rubber material is combined with neat bitumen and elemental sulfur to create the sulfur rubber bitumen binder composition. This combination was effective to increase the softening point of the composition as compared to the softening point of the neat bitumen.

Sylvester (2006)[31] applied sulfonic acids in rubber modified bitumen and their process of production to improve the properties of bitumen. The composition may require heat and/or mixing during or after addition of the sulfonic acid. The sulfonic acid may be linear sulfonic acids, branched sulfonic acids, dodecylbenzene sulfonic acid (DDBSA or DBSA), tridecylbenzene sulfonic acid (TDBSA), 4-methylbenzenesulfonic acid, dimethylbenzene sulfonic acid, toluene sulfonic acid. The rubber modified bitumen may be heated to a temperature range 225 °F to about 450 °F.

2.2.1 HISTORY OF WET PROCESS

The use of crumb rubber for bitumen modification in paving roads has been started in the 1840s. The first experiment was conducted by using natural rubber and bitumen. Due to elastic properties of natural rubber, it was used for the modification of bitumen to get more wear-resistant bitumen. Since 1960, shredded waste tires are also recycled for modification of bitumen.[32] Wet process of CRMB mixture is mixed for at least 45 min at a temperature of 180–210 °C and then there is a maturation of rubber-modified bitumen for 3 h at 180 °C without stirring.[33]

In mid-1980's, a special attempt were initiated to reduce the pollution caused by burning of scrap tires. Due to these fires air pollutants, oils, soot, and other materials are generated which contaminated air, water and soil. The use of scrap tire rubber in bitumen has been suggested as a potential partial solution to this environmental solid waste problem and it may enhance pavement performance. The addition of crumb rubber to bitumen, contributes not only to solve the problem of utilization of waste tires, but also affects the improvement of the mechanical properties of bitumen.[8]

The wet process of CRMB was first developed by Charles H. McDonald (1981)[34] for the modification of bitumen using fine crumb rubber at a specific temperature. The schematic diagram of McDonald's wet process is given in (Figure 2.1). He proved that rubber modified bitumen is characterized by the favourable characteristics from both the base materials. Moreover, the use of crumb rubber into bituminous mixtures needs a special care as rubber interacts with bitumen at high temperatures and can be changed the performance of the bituminous mixtures.[15, 35] At the same time, storage stability is also another main issue related to CRMB at high temperature. Thus, it is crucial to empathize the fundamental interaction of crumb rubber in bitumen.

2.3 INTERACTION OF BITUMEN WITH CRUMB RUBBER

Crumb rubber is synthesized using both vulcanized and synthetic rubber having with three-dimensional cross linked molecular structure.[36,37] The degree of swelling of CR in bitumen depends upon the content of solvent (maltenes portion) in bitumen.

2.3.1 OVERVIEW OF THE BITUMEN-CRUMB RUBBER INTERACTION

Mechanistic study of bitumen and CR interaction has not evaluated in detail. It is noted that the interaction of bitumen–rubber is not fully chemical in nature.[15]

This interaction occurred by two simultaneous processes (Figure 2.1): one is partial digestion of the CR in bitumen and another is absorption of oils from the bitumen by network structure of rubber, to swell and soften.[38] There is a formation of gel like material inside the body of bitumen when the rubber particles are swollen by absorbing oil portion of the bitumen at high temperature. Due to this absorption process the sizes of swollen rubber particle are increased with a continuous diminution of the inter-particle distance between them. Thus, throughout the reaction, a constantly decrease in the oily portion of bitumen to form gel structures can cause an in increase in viscosity of bitumen.[15]

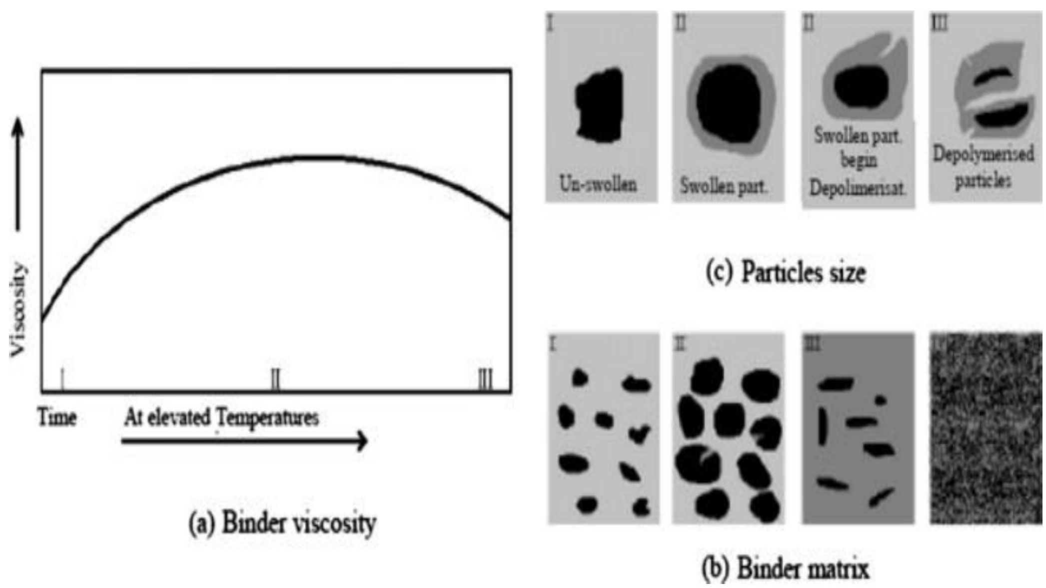

Figure 2.1: Bitumen-crumb rubber interaction phenomenon at elevated temperature over time[39]

According to researcher, the rate of swelling depends mainly on temperature and size of crumb rubber particle [36, 40-41].

2.3.2 EFFECT OF TEMPERATURE

Flory and Rehner (1943)[36] observed that, there are two effects of temperature on swelling of rubber in bitumen without changing in the composition of the rubber. An effect is the rate of swelling increases and other is extent of swelling decreases with increased temperature. There is an increase in elastic force and decrease in diluting force when more and more solvent enters to the rubber network. A state of equilibrium swelling is reached when these two forces become equal. However, more solvent can enter to the rubber particle when the temperature increases and then maintained.

Green and Tolonen (1977)[40] demonstrated that the reaction occurs above 155 ^{0}C and found that, the volume of swelling increases as temperature increases due to throwback of the rubber network. Due to this reversion, the cross-link density as well as requirement of entropy for expansion of the rubber network is decreased. Similarly, Singleton (2000)[35] used flux oil for swelling of crumb rubber particles at different temperatures and obtained that swelling increases correspondingly with temperature.

2.3.3 EFFECT OF PARTICLE SIZE

The physical properties of modified bitumen-crumb rubber blend are determined by the particles size distribution of crumb rubber. The interaction mechanism on swelling of the crumb rubber constituent part by absorption of light fractions present in the binder. Thus, the free space between the rubber particles is limited in swelling process, which constricted the rubber particles in their movement into the binder matrix. The finer particles as compared to the coarser particles swell easily providing higher binder modification.[42]

D. Lo Presti (2013)[32] also showed that a stable rubber modified bitumen can be obtained by using very fine rubber particles (size below 0.3 mm), and the modification process must be carried out at very high temperatures (200–300 °C) with a very high shear stress during the mixing of bitumen with rubber (mixing speed up to 8000 RPM) and at a pressure higher than 1 atm. In this condition, very fine rubber particles are devulcanized or digested in the bitumen matrix. So, the phase separation is not observed in the system. This system is referred to as No-

Agitation Recycled Tire Rubber Modified Bitumen (RTR-MBs) or terminal blended binder. There is no need to mix the system during storage. Unfortunately, RTR-MBs have worse final properties compared to the conventional rubber modified bitumen obtained through standard wet process from rubber particle with large particle size. Thus, the best efficiency of bitumen modification is achieved when the rubber particles are only partly swollen by the bitumen.

2.4 IMPROVEMENT FOR STORAGE STABILITY OF CRMB

The physico-chemical properties of modified-bitumen are better compared to unmodified bitumen. Generally, copolymer like SBR, SBS are mostly used to modify bitumen and crumb rubber (CR) obtained from scrap tires are used as a bitumen modifier due to its low cost. These polymers improve the property of bitumen i.e. rutting resistance, fatigue resistance and thermal cracking etc. However, there is a major problem in polymer-modified bitumen is low resistance to aging and storage issue at high temperature. Several methods are explored for the improvement of storage problem of CRMB.

V. Gonzalez et al. (2012)[43] studied the properties of bitumen modified with CR and a polymer to improve storage issue. They found that using polymeric additives in bitumen with improved mechanical and rheological characteristics of the binder.

B. Zhang et al. (2009)[44] demonstrated the effect of SBR and montmorillonite (SBR/MMT) for bitumen modification. X-ray diffraction results indicated that, there may be creation of an intercalated structure by SBR/MMT composites. The morphological properties of the samples were carried out using fluorescent microscopy and found that formation of fine structure in SBR/MMT modified bitumen. The SBR/MMT modified bitumen enhanced the SP viscosity with reduced penetration. The result confirmed that storage stability of modified bitumen is SRB/MMT content range dependent.

S Wang et al. (2014)[45] used like (HDPE) high-density polyethylene for the stabilization of bitumen by screw extrusion. They studied the effect of screw extrusion on storage stability and rheological properties of CRMB. They found that the storage stability of HDPE and CRMB was significantly upgraded after screw extrusion and HDPE and CR particles were more uniformly dispersed in the bitumen with improved rheological properties.

C. Oyuang et al. (2012)[46] used low-density polyethylene (LDPE) with maleic anhydride-grafted ethylene-octene copolymer (POE-g-MA) for bitumen modification. The toughness value was achieved to the highest level with softening point higher than 60 °C with amount of POE-*g*-MA was (8 %), and the softening point reached at 82 °C when the blend content was (8.5 %).

S. Kocevski et al. (2012)[47] characterized the surface modifications of ground tire rubber (GRT) by grafting acrylic acid without using any initiator for paving applications. The micro structural and morphological studies of the GRT particles were analysed by SEM and ATR-FTIR. The dynamic shear rheometer (DSR) test, thermal gravimetric analysis (TGA) viscosity test of the modified bitumen binder was investigated and found that there is increasing in the failure temperature and viscosity values of bitumen binder by the adding surface treated GRT constituents.

Ghaly, N. F. (2008)[9] explained the improvement of storage stability of CRMB using styrene-butadiene-styrene (SBS) and elemental sulfur (S). They have informed that storage stability of modified and unmodified bitumen was estimated at 140, 116 and 200 °C for critical vulcanization temperature. They found that storage stability of SBS modified bitumen after vulcanization process has been achieved at 160 °C while the critical degradation temperature reached to 200 °C, with increased Marshall Stability and resistance to plastic deformation with addition of sulfur with a combination of (4%) SBS and (1%) S.

Liu et al. (2014)[48-50] used 4% of trans-polyoctenamer rubber (TOR) in the modification of storage stability of rubberized bitumen. There was a formation network structure of polymer chains due to reaction of unsaturated bonds of TOR may react with sulfur from asphaltenes as well as sulfur present on the surface of ground rubber. Furthermore, rubberized bitumen with TOR has a lower decrease in viscosity in comparison with rubberized bitumen without TOR under storage for 24 h at a temperature of 177–190 °C which shows that the positive effect on improving the workability of rubberized bitumen.

Memon (1998)[51] demonstrated in his patent, the use of hydrogen peroxide and special compatibilizers for the modification of rubberized bitumen. In this modification process, the GTR is mixed with hydrogen peroxide at room temperature, and then heated to a temperature of 65–75 °C to generate carbonium ions on the surface of GTR grains. Then, the bitumen is heated to a temperature of 160–170 °C and mixed with a compatibilizer, which creates the bridges between bitumen and GTR grains. The compatibilizer may be methacrylate with glycidyl containing monomer, terpolymer of ethylene, butyl acrylate and glycidyl methacrylate or ethylene based glycidyl containing monomer. There is a better dispersion of GTR in bitumen by using special compatibilizers with improved rheological properties at both low and high temperatures.

S. Biro et al. (2007)[52] patented a method to improve storage stability of CRMB using several modifiers. First, bitumen was mixed with rubber granulate (3–40 wt. %, grain size below 2 mm) at the temperature of 70–230 °C. Then the modifiers were introduced to rubber bitumen mixture as A (oleophile hydrocarbon group, 0.03 wt. %), B (polyolefins, 0.01–4 wt. %) and C (amines, 0–2 wt. %). After adding all the components, the blend was stirred initially for 0.5–4 h, at the temperature of 200–270 °C and then next for 0.5–6 h, at 160–190 °C. The obtained binder was stable during storage at high temperature. They have claimed that it was, there is a formation of bonds between bitumen molecules and rubber granulates due to addition of the modifier A.

Perret et al. (1999)[53] indicated the improvement of stability of rubber modified bitumen containing (7–13 wt. %) of rubber granulate with particle size between 0.01 and 1 mm by using special kind of copolymer (1–2 wt. %) was added to rubberized bitumen. The copolymer was synthesized by reaction of alphaolefin with unsaturated epoxides. They had concluded that the rubberized bitumen modified by the copolymer was stable after 3 days of storage at 180 °C and it softening point was higher by about 13–20 °C while penetration value decreased by 15–30 [1/10 mm] compared to non-modified rubberized bitumen.

Shatanawi et al. (2012)[10] exhibited a method for improving stability of rubberized bitumen by using CR activated with 2-furaldehyde (C5H4O2). They explained the modified CR has higher reactivity and greater interactions between the CR and bitumen. Furthermore, it was concluded that the bitumen modified by CR (ambient grinding) was more stable than bitumen modified by CR (cryogenic grinding).

Moran (1991)[54] patented a method to advance the storage stability of modified bitumen using a polymer which has been pre-treated with an inorganic acid. The acid treated polymer modified bitumen was used as a binder in paving materials. The selected acid may be hydrochloric acid, phosphorus pentoxide, sulfuric acid, phosphoric acid and mixtures thereof.

Martin (2011)[55] depicted a method of modifying bitumen with waste rubber, and polyphosphoric acid. In this patent the mixture of bitumen, crumb rubber (0.5–25 wt. %) and polyphosphoric acid (0.05–5 wt. %) was rapidly stirred at 163–288 °C. The modification process was carried out for 1–24 h with blowing air through the system to enhance storage stability.

Formela and Maslanka (2013)[56] patented a method improving stability of rubber modified bitumen by using twin screw extruder wherein the extruder screws are co-rotating or counter-rotating (the temperature of cylinder 60–400 °C). The compositions contained bitumen grade 160/220 (max. 99 wt. %), ground rubber (1–99 wt. %; grain diameter<5mm), and additives (compatibilizer, modifiers, plasticizers, max. 80 wt. %). According to them, obtained binders had better stability in comparison to binders obtained through traditional method.

S. Wang et al. (2015)[57] established a way to improve storage stability of bitumen by using ground tire rubbers (GTRs), polyethylene and thermoplastic elastomers (TPEs). TPEs were prepared by dynamic devulcanization (DD) with desulfuriser and dynamic vulcanization (DV) with sulfur respectively. They have found that the physical properties were decreased by DD process, while strengthened of TPE by DV process.

T. Ma et al. (2015)[58] studied the modification mechanism with performance properties of the desulfurized rubber bitumen and CRMB. They found that, the performance of bitumen binder and mixture can be improved by both CR and desulfurized rubber. But the storage stability of desulfurized rubber bitumen is more than the crumb rubber bitumen.

Yetkin Yildirim (2007)[12] showed that SBR, SBS, EVA, polyethylene, Elvaloy, Ground tire rubber (GTR), elastomer, plastomers were the most preferable modifiers for bitumen modification to improves its properties, since, SBS copolymer in modified bitumen absorbs the oil fractions from bitumen and swells up resulting in three phase structures. Similarly, they have studied that, Elvaloy modifier helped to increase the storage stability of modified bitumen during transportation and application with improved performance properties at high temperature by making a chemical bond with bitumen.

F.J. Navarro et al. (2004)[59] studied the mechanical characteristics, storage stability and rheological properties of modified bitumen by 9 wt. % crumb tire rubber. They found that the accumulation of crumb rubber in bitumen increased the viscosity at high temperatures, which reduced the storage at low temperatures, which in turn resulted a greater flexible binder. They also found that crumb rubber of particle sizes <0.35 mm with high shear rates have high storage stability and decreased with increased in particle size and temperature and only 0.29 mm size rubber was stable under storage conditions.

Martin (2008)[60] used crumb rubber, one or more acids and a cross-linking agent for bitumen modification. The acid may be phosphoric acid, polyphosphoric acid (more than 100% expressed as orthophosphoric content), sulfuric acid, boric acid, and carboxylic acids and the cross-linking agent is elementary sulfur. This modified bitumen showed an improvement in elastic behaviour of binder.

Partanen (2010)[61] depicted a way to improve storage stability of recycled tire rubber modified bitumen by adding carbonaceous waste solids derived from petroleum refinery operations. Firstly, the carbonaceous waste solids were subjected to a heat treatment to release volatile materials and then combined with bitumen composition to achieve storage stable and of uniform consistency modified bitumen. The carbonaceous waste solids and recycled tire rubber having dimension of maximum of about 1 mm. Optionally, an oil selected from crude oil, a liquid petroleum fraction, motor oil, tall oil, agriculturally-derived oil, and clarified slurry oil was used. The heat treatment of mixture was maintained at a temperature of from about 100 °C to about 260 °C for a duration of 30 minutes to about 24 hours to release moisture and volatile organics from recycled tire rubber and thereby cause a reduction in weight of said mixture.

Zhigang et al. (2005)[62] used desulfurized crumb rubber (DCR) with reactive additives like sulfur and diisopropylbenzene peroxide for bitumen modification. They showed that not only the storage stability but also softening point and elasticity recovery of DCRMB were improved.

Literature reported regarding storage stable crumb rubber modified bitumen shows no prior literature is available on the anchoring of crumb rubber. Therefore, present study is focused on to improve storage stability of CRMB through chemical anchoring. Amines and amide moieties are known to react very quickly in bitumen matrix. Therefore, we have taken long chain amines and amide additives for CRMB anchoring. Furthermore, use of waste PET amide derivative also provides an ecofriendly way for disposal of waste plastic and produce CRMB with high storage stability. Accordingly, following novel additives with active functional groups have been investigated.

- Use of long chain amine compounds to form a storage stable CRMB.
- Use and study the synergic effect of polyamine with fatty acid in modification of crumb rubber modified bitumen.

- Use of synthesized waste PET derivative along with a bifunctional compound for anchoring the CRMB.

The CRMB prepared in the present research work not only storage stable but also possesses improved physical, rheological and performance properties.