

Use of Sulphur as an Additive in Bitumen: A Review

Dr. Maninder Singh¹, Dr. Kunal Jain², Sukhjinder Singh Kahlon³

¹Assistant Professor, Department of Civil Engineering, Punjabi University Patiala

²Assistant Professor, Department of Civil Engineering, Punjabi University Patiala

³M.Tech Scholar, Transportation Engineering, Punjabi University Patiala

Abstract - The increased volume of traffic creating higher stresses and temperature changes influencing binder efficiency to withstand distresses usually occurs in the form of fatigue cracking and permanent deformation in the pavement. To rectify these hurdles, adding additives to normal bitumen disseminates better engineering properties and helps to increase the bituminous pavement's life span. Sulphur is one of the pre-eminent additives used in the construction industry in various forms such as sulphur as a modifier, extender and a cross-linking agent. Elemental sulphur's distinct physical and chemical properties allow it to be used as a construction material. This paper reviews the researches carried out on sulphur as an additive. In view of previous studies, the benefits of using sulphur as an additive have been studied.

Key Words: Sulphur, Modified Bitumen, extender, cross linking agent

1. INTRODUCTION

During the 1930s, in Texas first comprehensive studies on the treatment of various sulphur asphalts at relatively lower temperatures were conducted. The impact of sulphur on the bitumen and mixtures was stated by (Benzowitz and Boe, 1938). Then, attempts were made by the US Bureau of Mines and Federal Highways (Kennepohl et al., 1975; Kandhal, 1982) during the 1970s until the early 1980s to consider sulphur as an extender to decrease the quantity of asphalt binder included in mixtures and enhance the mechanical properties of the mixture. (Timm et al, 2009).

In the late 1990s, sulphur pastillation process was advanced, which simplified the operations of element sulphur in a solid-state. Further, with the introduction of plasticisers in sulphur pellets that allow the sulphur application to asphalt mixtures with substantially reduced pollutants and odour relative to sulphur asphalt mixtures in the liquid form. Through advancements in modified sulphur production in pellets form, enhanced handling and performance properties, created tremendous interest in bitumen modification (Jacques Colange et al, 2010).

Sulphur

Sulphur is a lemon yellow sintered microcrystal, having 16 protons and 16 electrons with 16 neutrons with the atomic weight of 32 atomic mass units and recognized by

the letter S. It is used to produce several goods for domestic, agriculture and many other purposes. Sulphur has a melting point of 388.36 Kelvin, or 239.38 degrees Fahrenheit and a boiling point of 717.8 Kelvin or 832.3 degrees Fahrenheit with a density of 2.07 grams per centimeters cubed. Compared with the old SEA technologies, the simplicity of processing and integration of modified sulphur pellets made it more convenient, especially during the mixing phase. Globally, China produces about 17.4 megatons of sulphur from natural gas and petroleum which made China the world's leading sulphur producer in 2019, whereas India ranked 9th with 3.4 megatons of sulphur production annually.

2. Literature Review

2.1 Sulphur Modified Bitumen

Jacques Colange et al (2010) investigated the sulphur modified asphalt mixtures its impact on the thickness of pavement, using a pavement design analytical method (BISAR). Materials such as bitumen and modified sulphur pellets used as well as laboratory tests such as empirical tests such as Marshall, durability, rutting resistance as well as high and low temperature properties were perforated. During efficiency experiments used to classify HMA, materials such as bitumen and customized sulphur pellets tested in laboratory. Fundamental properties, analytical pavement investigation as well as experiments such as empirical measures like durability of Marshall, rutting scales, low and high-temperature properties were conducted. Thus, the results obtained from tests depicts that modified sulphur pellets enhanced asphalt mixture properties with increased workability and leads to ease of compaction at mild mixing asphalt manufacturing operating temperatures and enhanced permanent deformation resistance for roads exposed to heavy loads. Furthermore, Low-temperature properties of varying sulphur quantities up to 40 per cent of the binder mass were equal to or even marginally stronger than standard asphalt mixtures.

Samuel B. Cooper III et al (2013) investigated the effectiveness of modified sulphur mix additive in warm-mix asphalt. For this, properties of sulphur-modified warm-mix asphalt (WMA) and unmodified asphalt mixtures (HMA) were evaluated. There were exactly 3 types of the mixture, one WMA and rest two of HMA. One

mixture was without any modification, classified as PG 64-22; the other two mixtures were modified with graded elastomeric styrene-butadiene-styrene binder named as PG 70-22. A series of tests performed to assess rutting efficiency, resistance to moisture, fatigue endurance, resistance to cracks and thermal cracking and others along with constrained specimen test with thermal tension. Consequently, the Loaded-Wheel Tracking, Repeated Shear at Constant Height Test and Flow Number tests revealed that the sulphur-modified WMA mixture was better to perform against rutting as compared to the conventional mixtures and polymer-modified asphalt binders. The findings of Thermal Pressure Restricting Specimen Test revealed that in WMA modified with sulphur had the fracture stress higher than that of bitumen modified with polymer. Nevertheless, findings of fracture experiments revealed that WMA had been modified by sulphur had more vulnerability to cracking in contrast with conventional mixtures due to its stiffness. Despite stiffness properties, the modulus of bitumen modified with sulphur was higher and minimized the magnitude of strain caused by the pavement.

Abdulgazi Gedik et al (2013) conducted a laboratory investigation, aims to examine the appositeness of sulphur as compare to neat bitumen and determine the practical application of the resulting bitumen modified with sulphur in flexible pavements. Materials such as neat bitumen binder (70/100 penetration grade) as well as sulphur-added bitumen (SAB) and aggregates of crushed quartz used in this research. After analysis, the Viscosity test explicated that the integration of sulphur into neat bitumen declined the mixing and the compacting temperature of binder course specimens. This process not only tended to minimize energy usage, but also a tendency to reduce energy use. Furthermore, from rheological observations, adding sulphur had a positive impact on composite binder as its inclusion created strong structure within bitumen matrix, imparted better performance and high temperature resistance. Besides, image analysis revealed, sulphur particles had no longer soluble in the case of a higher concentration and remained same in the bitumen phase of colloidal suspension. Next to this, SAB's composite mixture had improved Marshall Stability and retained its flow ability, significantly enhanced the pavement performance against the high-temperature deformation.

Dr Praveen Kumar and Nikhil Saboo (2014) investigated on the mechanical and rheological properties of the sulphur modified binder, in which sulphur added from 2-40% in bitumen and the improvement in properties were evaluated by both empirical tests and Dynamic Shear Rheometer. Also, concentrates on the requirement for sulphur modified binders, leading to the advancement of sustainable guidelines for pavement. VG-10 Viscosity grade binder, sulphur in pellet form were

used, in this analysis. Modification of the sulphur results in lower mixing and compaction temperature conditions, rendering sulphur-modified bituminous mixture a warm asphalt mixing technology and Sulphur serves as an extender and substitutes for bitumen, hence, protects extra bitumen from burning and further adds to environmental conservation. Bitumen alteration with sulphur enhances the plasticizing effect at lower modification levels. Inclined sulphur concentration (20 per cent by weight of total binder) increases binder's rigidity. Total energy consumption ascribed to lower paving costs as using coal to heat the bitumen at higher temperature leads to lower.

Poorna Prajna S and Mohamed Ilyas Anjum (2015) investigated the impact of sulphur colloidal powder as a modifier, comprised a mixture of 75% sulphur and 25% acacia (gum Arabic) as well as determined the Marshall properties of bituminous mixes by using bitumen grade of 60/70 penetration binder and sulphur modified bitumen. The bituminous Concrete mixtures correlating to grade-1, prepared by utilizing 30% combined index aggregate at midpoint gradation with sulphur as a modifier. As a result, the maximum value of Marshall Stability found to be 30.22 kN for 9 per cent sulphur at 5 per cent optimum bitumen content, which was higher than plain bitumen. The maximum bulk density for unmodified and modified bitumen was found at 3 per cent, 6 per cent and 9 per cent sulphur addition at 5.5 per cent bitumen material respectively. Air voids reduction leads to improvement in the strength and performance period of pavement and VFB had been increased by adding bitumen. The optimum binder content of and modifier found to be 5 per cent and 9 per cent respectively, as per MORTH. Consequently, modification of bituminous concrete mixes had resulted in greater stability with less bitumen material.

Dawid D'Melo et al (2016) examined the outcomes of elemental sulphur in a bitumen mixture with regards to time and sulphur influence on the properties of bitumen after modification. The bitumen selected for the analysis had a penetration grade of 160/220 and elemental sulphur was used in this research. As a result, the incorporation of sulphur with bitumen leads to creation of an amorphous sulphur phase that reconstructs itself over a period of time to create dendritic structures at concentrations of sulphur above 10%. The average sulphur content of more than 20 per cent results in crystalline sulphur phase occurred in a mixture that behaved likewise to bitumen filler. The creation of dendritic sulphur structures in sulphur modified mixtures relates to significant increase in stiffness of mixture. The sulphur modified bitumen mixture's stiffness observed to be increased over time. This enhancement in the stiffness of the sulphur modified bitumen found to be induced by observed amorphous sulphur overtime aggregation.

Aditya Kumar Das and Mahabir Panda (2017) evaluated the appropriateness of sulphur as a bitumen modifier for road construction by conducting the Marshall Stability Test on plain and sulphur modified bitumen specimens as per ASTM D 1559. In this study, the Marshall Test properties of bituminous concrete mixes using 60/70 bitumen penetration grade as well as sulphur and influence of sulphur as a modifier in different proportions of bituminous mixes was determined. The properties of sulphur modified bituminous like volume of air voids, the volume of bitumen, VMA, VFB, bulk density, theoretical density, flow, Marshall Stability, and Marshall Quotient values were analyzed. It was also observed, with the application of sulphur, VMA and volume of air voids were declined, whereas, theoretical density remains unchanged, however, values of Volume of bitumen as well as VFB were decreased. As a result, maximum bulk density was found to be 2.42 g/cc for normal and modified bitumen at 3 percent, 6 percent and 9 percent sulphur with 5.5 percent bitumen content, whereas Marshall Stability was found to be at the peak of 30.22 kN for 9 percent sulphur at 5 percent bitumen content. Bituminous modified mixes had resulted in utmost stability with less bitumen content, ultimately reduced the consumption of plain bitumen and to some extent, the cost of road construction.

Van Hung Nguyen and Van Phuc Le (2019) investigated the sulphur performance as an alternative binder's additive for Mixtures of asphalt. The engineering properties and morphology of the asphalt binder modified with sulphur were explored by using Electron Microscopy Scanning (SEM), Marshall Stability (MS) and other tests were used to assess the performance resistance of sulphur-modified asphalt mixtures. And then, the efficiency of sulphur-modified asphalt mixtures simulated and evaluated by using the MEPDG (Mechanical Analytical Paved Design Guide) program. Materials namely bitumen(60/70 grade) used as asphalt binder, yellow powder form sulphur in different percentages by weight of total binder and nominal greatest aggregate size of 12.5 mm dense gradation utilized with the sulphur modified asphalt mixtures. Results of the MEPDG program showed that by adding sulphur content to asphalt mixtures improved its rutting, cracking, and TD cracking under field conditions, performances considerably. Optimum 40 per cent sulphur material used as a substitution of bitumen binder, since, it enhanced the toughness, resistance to rutting, fatigue cracking and asphalt resistant mixtures.

Kumkum Priyadarsini and Jhunarani Ojha (2020) conducted experimental studies to determine the effect of sulphur as a modifier on the properties of binder along with ageing evaluation. Sulphur used as a modifier in the binder and its properties checked, as well as the effects of short-term ageing, was also determined. Sulphur had changed with varying percentages from 1 to 9 per cent and the physical properties of the modified binder tested by

different tests such as Ductility, Elastic Recovery, Viscosity, Penetration and Softening point. The optimum concentration of sulphur found to be 2%. Results showed that with rising percentages of sulphur, penetration and softening point values increased. However, the elastic recovery and ductility values fall and rose sequentially, with an increment in sulphur content. The effect of the ageing sulphur Modified binder was within the appropriate range and provides better results as opposed to pure bitumen.

2.2 Sulphur Extended Bitumen

G.H. Shafabakhsh et al (2015) scrutinized sulphur-extended asphalts modified with anti-stripping agents evaluated by using the Surface Free Energy method to check the moisture susceptibility. The specimens were prepared with two distinct aggregates such as calcareous and granite. The surface energy elements of constituent materials utilized for determining the bond strength among them in dry and wet conditions by using SFE method. Consequently, compatibility ratios (CR) revealed increased values for calcareous-mixes as opposed to granite-mixes. This indicates that calcareous aggregates were more defiant against moisture damage. Zycotherm as an anti-stripping agent strengthened the adhesion in mixture by rising and reducing adhesion strength, under dry and wet conditions respectively. The acid-to-base ratio was decreased with the addition of the anti-stripping agent (NZ) thereby made it more competitive with acidic aggregates. Although adhesive energy in mixture was higher as compared to calcareous in a dry state, calcareous stone displayed lower free adhesive energy in contact with mud, allowing calcareous mixtures more resistant to humidity relative to granite mixtures. Even though the sulphur was an acidic base element, it increased the acid component of the asphalt binder, while sulphur decreased the binder component (Lifshitz-Van Der Waals), thereby weakened the bond between the modified sulphur binder and acidic aggregates.

Vitaliy Gladkikh et al (2016) conducted modelling to determine the rutting kinetics of sulphur-extended asphalt. Sulphur-extended asphalt specimens prepared from diatomite powder of average particle size 7 μ m, gabbro-dabase chipping of fraction 5-20 mm, bitumen, granite screenings of fraction 0, 315-5 mm and sulphur pellets as a modifier for modelling of sulphur-extended asphalt's rutting kinetics. In this analysis, the additive parametric model described two instantaneous processes such as late densification and plastic flow to evaluate rutting kinetics. Consequently, Sulphur's rutting resistance extended asphalt by 1.3 to 3.7 times higher as compared to conventional asphalt concrete rutting resistance. The model's parameters determined based on the experimental data exhibited a monotonous decline in rate of plastic flow with increment in the quantity of sulphur

which indicates, high rutting resistance of extended asphalt of sulphur.

Masoud Faramarzi et al (2017) conducted several lab experiments to determine the mechanical properties and susceptibility to the moisture of Sulfur extended asphalt (SEA) mixtures modified with addition of anti-stripping agent (ASAs). To evaluate the efficiency, four types of mixtures of specific additive concentrations, including ASA and sulphur, were prepared and fatigue behaviour, resilient modulus, the susceptibility to moisture, rutting resistance tests were conducted. Modifying the asphalt mixture with Goo gas and using Zycotherm nanotechnology as anti stripping agent resulted in an efficient combination of additives that enhanced the mechanical properties of SEAs. At the one side, with an improvement in modulus of SEA mixtures, the severity of strain caused in sample was decreased (about 20%), and on the other side, the anti stripping agent improved fatigue behaviour (nearly 25%) by forming adhesion within asphalt and aggregates, albeit SEA mixtures experienced the least fatigue life. Scanning Electron Microscopy technique for mixing Goo gas into asphalt mixtures and creating an almost homogeneous matrix of Goo gas-asphalt shows successful results. Although several Goo gas components have been dissolved to an appropriate degree others have been crystallized at micro and nanoscales. Goo gas' expansion of a section of asphalt increases the resilient modulus of the mixture; as a result, it became more resistant to permanent deformation. At low and high temperatures, respectively, this rise was between 50 percent and 100 percent. Using ASA decreased the robust modulus of the SEA mixture significantly; this degradation was insignificant especially at higher temperatures (approximately less than 4 percent). According to Lottman's test examination, Goo gas' implementation decreased ITS and TSR by about 8 percent and 12 percent each; however, the ASA-modified SEA (ASWMA) moisture resistance had been greatly increased corresponding to unmodified SEA, as ASA substantially increased ITS and TSR by nearly 42 percent and 28 percent sequentially. Overall, the findings of this work suggest that the alteration of sulphur can boost the mechanical properties of asphaltic mixtures in rutting behaviour ultimately. Alternatively, Zycotherm nanotechnology strengthens the adhesion between SEA and aggregates.

M. S. Eisa et al (2019) evaluated the performance of low-quality aggregate hot asphalt mixtures with sulphur-extended asphalt. For this, a hot asphalt mixture of inferior quality aggregate and normal bitumen at 160 °C mixing temperature was prepared and many other mixtures of hot asphalt were made with poor quality aggregate along with commercial sulphur substitution (10%, 20%, 30%, and 40%) per cent as per weight of bitumen at 140 °C mixing temperature. Thereafter, several mechanical

parameters were calculated together with air voids, Marshall Stability, flow value, Stiffness and so on. Conclusively, the optimum sulphur replacement dose was 30 per cent by weight of bitumen along with inferiority aggregate, created better hot asphalt mixtures, resulted in decrease in bitumen consumption (21.4%), higher stability value (13%), modified flow value reduced to 15% and higher rutting resistance as compared to asphalt mixtures without adding sulphur. Asphalt mixtures containing inferior aggregates indicated that 30 per cent sulphur replacement showed a slight decrease in moisture Sensitivity with indirect tensile strength at 5.4% and slight rise in moisture sensitivity due to loss of stability at 9.6%, however, all results were within acceptable ranges compared to inferior asphalt mixtures without substitution of sulphur. In contrast, addition of sulphur in asphalt showed significant improvement in mechanical properties and rutting resistance.

2.3 Sulphur Modified Bitumen's Properties Compared with Other Additives

G. Fritschy et al (1981) analyzed the dynamic mechanical properties of binder modified with sulphur and fine silica particles in model composites. Bitumen and silica composites had dynamic viscoelastic properties, following principles of time temperature interfacial parameter equivalence. In the mixing procedure, both bitumen and sulphur (2%) were prepared at 140 °C for 30 minutes before being mixed and blended within 1 minute by using an effective stirrer. Consequently, chemical interaction of sulphur with bitumen formed a new binder, within which asphalt had a more gel-like structure consequential from higher aggregate-forming capability. There was a decrease in dynamic mechanical properties in normal binder, since, an evident plastification of the material; however, plastification seems not attributed to decrease the internal friction. As compared to transient systems of black carbon in elastomers, resembling was created by aggregates.

Abigail Martinez-Estrada et al (2010) conducted a comparative analysis of the impact of sulphur on SB and SBS-Modified asphalt morphology and rheological properties. This research analyzed the effect of asphalt alteration using styrene-butadiene block copolymers as well as sulphur to assess impact of polymer content, sulphur/polymer ratio and polymer molecular characteristics on physical properties of improved asphalt mixtures. Specific forms of styrene-butadiene copolymers (SB and SBS) were used varied greatly in recognition of chain composition, molecular weights, polystyrene blocks, the size as well as distribution of polybutadiene, were investigated by gel permeation chromatography, nuclear magnetic resonance and differential calorimetry scanning. A hot mixing procedure used to prepare synthetic asphalt blends, identified by binder tests, Rheology and so on. This research included materials such as Sulphur (99%), Mexican asphalt and deuterated chloroform, Styrene-

butadiene copolymers, Solprene S1205 and Solprene S416. Therefore, two different forms of styrene-butadiene copolymers with and without sulphur to assess effect on morphology and rheological properties of PMA like polymer concentration, molecular characteristics and sulphur/polymer ratio. Morphology of both polymer-rich process and its rheological actions with the application of small quantities of sulphur to PMAs have changed significantly, thus prevents phase separation during hot storage.

Feng Zhang et al (2010) studied physical properties and rheological changes in asphalt, modified by styrene butadiene styrene block copolymers and asphalt modified with sulphur by using a short-term and long-term ageing test, to compare asphalt modified with SBS against and sulphur modified asphalts by ageing, to explore impact of ageing on morphology of modified binders by optical microscopy. Also, results demonstrated issues that occurred in the practical implementation of SBS and sulphur-modified asphalts in storage. SK-90 paving asphalt, SBS1301 and SBS4303 (where former was star-like polymer whereas latter is linear polymer, both containing 30 wt% styrene) and Sulphur were used as ingredients. The dynamic rheological analysis verified the findings of viscosity and softening point just to a slight extension along with an illustration of two main causes of short and long term oxidative ageing on the rheological property of modified asphalts. At the one side, ageing induced polymer deterioration and increased the viscous behaviour, on the other side; ageing of the modified binders altered the structure of the asphalt as well as improved the elasticity of modified binders. Storage-stable SBS, as well as sulphur-modified asphalts, exhibited an apparent viscous behaviour as compared to asphalt modified with SBS that increased the creep quality at low temperature. Regardless of its composition SBS and sulphur-modified asphalt instability, SBS-modified asphalt rutting resistance declined by the addition of sulphur. Until the modified binders had the rheological properties and after ageing, the structural characteristics of SBS still relied heavily upon. Optical microscopy exhibited improvements in the compatibility of asphalt and SBS with more ageing.

M. Anwar Parvez et al (2013) used the waste crumb rubber 1 to 6 per cent and sulphur at different proportions from 20 to 50 per cent aimed to enhance the efficiency of asphalt binder in pavement applications. Thermal analysis, dynamic or steady shear rheology and artificial ageing techniques were used to investigate the melt properties. The ARES rheometer used to find the rheological properties of samples. Strategic highway research program factor ($G^*/\sin\delta$) and zero shear viscosity observed with an increase in crumb rubber percentages for each asphalt binders extended by sulphur which reflects the modification of crumb rubber, possibly

improved resistance against rutting at higher temperatures. The crumb rubber enhanced the sulphur-extended asphalt binder's viscoelastic properties checked by dynamic and steady shear rheology tests. Crumb rubber modification lowered sulphur and asphalt's temperature sensitivity and elevated sulphur asphalt upper grading performance temperature. The reduction in the energy of activation had been found due to combined effect of sulphur and crumb rubber as opposed to pure asphalt. Short-term ageing enhanced $G^*/\sin\delta$ with a slight increase in energy from activation. Incorporation of sulphur with asphalt mixture enhanced the viscoelastic properties of sulphur asphalt. Adding rubber crumbs to sulphur asphalt, increased resistance against temperature, further, use of crumb rubber and sulphur in asphalt had been demonstrated improvement in asphalt pavement life.

Íñigo Aguirre de Carcer et al (2014) studied the SBS changed bituminous stability behaviour in the presence of sulphur. This research conducted for objectives, to examine the structure and compositional effect on the stability of the bitumen and test the improvement in properties of SBS bitumen mixtures formed by storage at a high temperature of 160 degree Celsius. Various tests such as softening point, viscosity, and morphology of PMB samples conducted. Bitumen from various sources of crude oil had been used to prepare modified SBS/sulphur asphalt. Although bitumen used for this work revealed likewise values in their macroscopic physical properties, suitable for changes in polymer, and similar asphalt structures, but demonstrate evident differences in stability behaviour when a radial. Significant modifications in the aromatic material and the percentage of a solid phase (solid resin asphalt) were omnipresent. The action that was responsible for dispersed in maltene matrix, for strong modified-bitumen, required SBS interaction. Aromatic presence in these preparatory conditions was approximately 40 wt per cent content and solid phase volume, was below 22 per cent.

E.R. Souaya et al (2015) The analysis involved transforming elemental sulphur into more stable modified binder by utilizing combination of olefin hydrocarbon products, produced from petroleum fractional distillates and bituminous residue of cyclic hydrocarbon. To check out their morphology and structural characteristics of prepared modified sulphur, Nanoindentation, Structural analysis and scanning electron microscope (SEM) tests were conducted. Differential scanning calorimetric curves analysed to find the variations in sulphur phases from a-orthorhombic to b-monoclinic structure. After portrayal and clarification of asphalt modified with sulphur by using FTIR, XRD, SEM and DSC experiments, chosen residue mixture (C5 fractions and bitumen) obtained from petroleum distillate was used as an additive to produce stable polymeric sulphur. Probing further, Nanoindentation of normal and modified sulphur

exhibited, the modified sulphur had a greater mechanical resistance than plain molten sulphur. In hot mixture of Marshall Design; as far as the percentage of substituted sulphur increased, likewise, the Marshall Stiffness increased accordingly.

Ming Liang et al (2017) examined the effects of polymerised sulphur on morphology, stability of modified asphalt SBS and rheological properties. The rheological consequences were interpreted by the results from fluorescence microscopy and FTIR. However, dynamic shear modulus and viscosity evaluated for testing storage stability. The material utilised for this experimentation, styrene (30% by weight) with SBS linear polymer and 80/100 penetration grade asphalt used as base asphalt. The elemental sulphur noted as S0, there were total 3 types of polymerized sulphur used in this study noted as S40, S60 and S90, sequentially, in which, S40 contains 40% polymerized sulphur weight and remaining 60% was elemental sulphur. Concentrations of polymerized sulphur in S60 and S90 were 60% and 90% accordingly. As a result of the aforementioned tests, in microscopy observations, it had been shown; SBS phase domain vulcanized by polymerized sulphur was larger as compared to elemental sulphur. The domain continued to extend with increase in polymerized sulphur, indicating lower degree in vulcanization and density of cross-linking network. Dispersed SBS phase formed after transformation from large SBS phase progressively. FTIR results depicted that decrease of unsaturation of poly butadiene block and creation of CAS bond was major evidence of vulcanization. Due to extension of mixing time, complex viscosity of SBS modified asphalt along with sulphur performed better than to unmodified sample. In contrast, rise in dynamic viscosity of polymerized sulphur was less quick than elemental sulphur and further became slower with increase in the quantity of polymerized sulphur. Likewise rise in viscosity at 135 C with time was also detected, which implies that vulcanization rate falls for polymerized sulphur samples. Polymerized sulphur leads to a greater rise in steady viscosity, complex viscosity and fluid viscosity at 135 C of SBS changed asphalt, relative to a sample made in the absence of sulphur. Nevertheless, such viscosity functions declined with increased polymerized sulphur concentrations in functions of viscosity in samples of elemental sulphur were higher than polymerized sulphur. Therefore, polymerized sulphur revealed, inferior effectiveness of vulcanization on SBS in contrast with elemental sulphur and lower amount of sulphur correlates to lesser vulcanization performance.

Naipeng Tang et al (2017) examined the rheological analysis of the terminal mixture of rubberized asphalt binder with polymeric addition and sulphur. For this analysis, hybrid asphalt binders of Terminal Blend (TB)

including crumb rubber, styrene-butadiene-styrene (SBS) and sulphur prepared at different proportions were prepared. Storage stability test, multiple stress creep recovery test and other tests were conducted to determine rheological characteristics of Terminal Blend hybrid binders. The maximum rheological curve developed at 25 °C used to differentiate TB hybrid binders' rheological behaviour over range of frequencies. The findings showed that SBS and sulphur improved higher temperatures and elastic properties of TB rubberized asphalt binders. Fewer M-controlled TB hybrid binders made with an increase in the amount of sulphur. It was recommended and mentioned through laboratory studies, the production of TB hybrid binders from 0.2 to 0.3 per cent with acceptable storage capacity, appropriate viscosity and balanced efficiency at high and low temperatures.

Wachira Saowaparka et al (2017) investigated the storage stability, physical properties, morphology and rheology of natural rubber (NR) modified asphalt, polyphosphoric acid as well as sulphur. 60/70 penetration grade asphalt, latex form Natural Rubber used in this analysis. Consequently, It was found that inclusion of 0.3 wt per cent of sulphur as per Natural Rubber content in NR/PPA-modified asphalt considerably increased toughness and tenacity of asphalt obtained from effect of vulcanization. These aforementioned modifiers provided complementary effects in the improvement of asphalt properties. The modified asphalt with 3.2 wt per cent natural rubber was 2 wt per cent of the above results. The optimal formula for producing high stability asphalt mixture for road pavement applications was indeed 0.3 wt per cent of sulphur (based on 100 parts NR).

Ashlyn D. Smith et al (2020) combined the elemental sulphur, oleic acid and either OPC or pozzolan cement and a range of six sulphur cement pastes to prepared and determine the effect copolymerization. These pastes were not only equipped mainly from waste or viable materials but also showed significant improvements in acid resistance as compared to conventional OPC mixtures. Blocks originated from aforementioned pastes had several prominent characters of commercial sulphur cement with extra desirable characterisation of excessively lower water absorption, thermal healing property and higher mechanical resistance after exposure to strong oxidizing solutions of acid than conventional Portland cement. Surface damage to blocks of sulphur cement was healed by thermal annealing at varying extents, as assessed through optical microscopy.

3. CONCLUSIONS

Sulphur as an additive in asphalt mixes has been adapted for better structural enhancement of the pavement by enhancing material properties as well as help to preserve natural resources. Bitumen modification with sulphur enhances penetration, stiffness, viscosity of binder and

softening point values. Not only this but also the sulphur modification contributes to lower the compaction temperature and reduces mixing requirements. Moreover, the Marshall Stability value, voids filled with Bitumen and the bulk density improved, however, air voids reduced with the introduction of sulphur in bitumen. Furthermore, sulphur bitumen modification increased the Aging index value, thus reduced the binder's ageing susceptibility. Elastic recovery and ductility values rose and fall respectively, with the usage of sulphur content. Comparatively, Sulphur-modified in Warm Mix Asphalt had almost likewise resistance to moisture, rutting performance and susceptibility to cracking than polymer-modified as well as unmodified asphalt binders. Conclusively, the performance of the SMB bitumen, based on the reaction of rutting, fatigue, ageing and storage stability, makes sulphur an effective bitumen modifier. Apart from this, in sulphur extended asphalt, the plastic flow rate declined with an increase in sulphur quantity, which indicates high rutting resistance.

REFERENCES

1. Benzowitz, J. and Boe, E.S., Effect of sulfur upon some of the properties of asphalt. Proc. ASTM, 38: 539 (1938).
2. Kandhal P.S., "Evaluation of Sulphur Extended Asphalt Binders in Bituminous Paving Mixtures" Proceedings of the Association of Asphalt Paving Technologists, Vol. 51, 1982, p. 189-221.
3. Kennepohl G.J.A., Logan A. and Bean D.C., "Conventional' Paving Mixes with Sulphur- Asphalt Binders", Asphalt Paving Technology, Proceedings, Association of Asphalt Paving Technologists, Technical Sessions, Phoenix, Arizona, February 10-12, 1975, p. 485-518.
4. Timm, D., Tran, N., Taylor, A., Robbins, M., and Powell, B. (2009). "Evaluation of mixture performance and structural capacity of pavements using Shell Thiopave, Phase I: Mix design, laboratory performance evaluation and structural pavement analysis and design." NCAT Rep. 09-05, National Center for Asphalt Technology, Auburn, AL.
5. Jacques Colange, David Strickland, Gordon McCabe, Kevin Gilbert, Richard May, Simon Banbury (2010), Asphalt Mixtures Modified with Sulphur Pellets Impact on Pavement Thickness. Road Materials and Pavement Design, 11:sup1, 459-485. DOI: 10.1080/14680629.2010.9690342
6. Samuel B. Cooper III and Louay N. Mohammad (2013), Laboratory Performance Characteristics of Sulfur-Modified Warm-Mix Asphalt Vol. 23, Issue 9 (September 2011). DOI: 10.1061/(ASCE)MT.1943-5533.0000303.
7. Abdulgazi Gedik and Abdullah Hilmi Lav (2013), Sulphur Utilization in Asphaltic Concrete Pavements, 2013 Airfield & Highway Pavement Conference, DOI: 10.1061/9780784413005.099
8. Dr. Praveen Kumar and Nikhil Saboo (2014), Rheological Investigations of Sulfur Modified Bitumen. ICSCI 2014 ASCE India Section, Oct 17-18, 2014, Hitex, Hyderabad, Telangana, India.
9. Poorna Prajna S and Mohamed Ilyas Anjum (2015), Suitability Of Sulfur As Modifier In Bitumen For Road Construction. International Journal of Research in Engineering and Technology, eISSN: 2319-1163, pISSN: 2321-7308.
10. Dawid D'Melo, Sridhar Raju, Subhendu Bhattacharya and Sathish Subramani (2016), Self-assembly of amorphous sulphur in bitumen-sulphur mixtures and its impact on properties. Construction and Building Materials 126 (2016) 976-982 <https://doi.org/10.1016/j.conbuildmat.2016.09.114>
11. Aditya Kumar Das and Mahabir Panda (2017), Investigation on rheological performance of sulphur modified bitumen (SMB) binders. Construction and Building Materials 149 (2017) 724-732. DOI: <https://doi.org/10.1016/j.conbuildmat.2017.05.198>
12. Van Hung Nguyen and Van Phuc Le (2019), Performance evaluation of sulfur as alternative binder additive for asphalt mixtures. DOI: <https://doi.org/10.1007/s42947-019-0045-9>.
13. Kumkum Priyadarsini and Jhunarani Ojha (2020), Experimental Studies On Properties Of Sulfur Modified Binder With Aging. International Journal of Civil Engineering and Technology, 11(2), 2020, 140-155. <http://www.iaeme.com/IJCIET/index.asp>
14. G.H. Shafabakhsh, M. Faramarzi and M. Sadeghnejad (2015), Use of Surface Free Energy method to evaluate the moisture susceptibility of sulfur extended asphalts modified with antistripping agents. Construction and Building Materials 98 (2015) 456-464. DOI: <https://doi.org/10.1016/j.conbuildmat.2015.08.123>.
15. Vitaliy Gladkikh, Evgeniy Korolev, Vladimir Smirnov and Ilya Sukhachev (2016), Modeling the rutting kinetics of the sulfur-extended asphalt. Procedia Engineering 165 (2016) 1417 - 1423. DOI: <https://doi.org/10.1016/j.proeng.2016.11.873>
16. Masoud Faramarzi, Behnam Golestani and K. Wayne Lee (2017), Improving moisture sensitivity and mechanical properties of sulphur extended asphalt mixture by nano antistripping agent. Construction and Building Materials 133 (2017) 534-542. DOI: <http://dx.doi.org/10.1016/j.conbuildmat.2016.12.038>
17. M. S. Eisa, M. E. Basiouny and O. A. Elbasomy (2019), Evaluating hot asphalt mixtures of poor quality aggregate with sulphur extended asphalt. Innovative Infrastructure Solutions (2019). DOI: <https://doi.org/10.1007/s41062-019-0241-0>
18. G. Fritschy, E. Papirer and C. Chambru (1981), Sulphur Modified Bitumen, A New Binder Rheologica Acta, Vol.

- 20, No. 1 78-84 (1981), ISSN 0035-4511, ASTM Coden : RHEAAK.
19. Abigail Marti'nez-Estrada, A. Enrique Cha'vez-Castellanos, Margarita Herrera-Alonso and Rafael Herrera-Na'jera (2010). Journal of Applied Polymer Science, Vol. 115, 3409-3422 (2010), DOI: 10.1002/app.31407.
20. Feng Zhang, Jianying Yu and Shaopeng Wu (2010), Effect of ageing on rheological properties of storage-stable SBS/sulfur-modified Asphalts. Journal of Hazardous Materials 182 (2010) 507-517, DOI:10.1016/j.jhazmat.2010.06.061.
21. M. Anwar Parvez, Mohammed Al-Mehthel, Hamad I. Al-Abdul Wahhab and Ibnelwaleed A. Hussein (2013), Utilization of Sulfur and Crumb Rubber in Asphalt Modification. J. APPL. POLYM. SCI. 2014, DOI: 10.1002/APP.40046
22. Íñigo Aguirre de Carcer, Rosa M. Masegosa, M. Teresa Viñas, Marta Sanchez-Cabezudo, Catalina Salom, Margarita G. Prolongo, Verónica Contreras, Francisco Barceló and Antonio Páez (2014), Storage stability of SBS/sulfur modified bitumens at high temperature, Influence of bitumen composition and structure. Construction and Building Materials 52 (2014) 245-252, DOI: <http://dx.doi.org/10.1016/j.conbuildmat.2013.10.069>.
23. E.R. Souaya, S.A. Elkholy, A.M.M. Abd El-Rahman, M. El-Shafie, I.M. Ibrahim and Z.L. Abo-Shanab (2015), Partial substitution of asphalt pavement with modified sulphur. Egyptian Journal of Petroleum (2015) 24, 483-491, DOI: <http://dx.doi.org/10.1016/j.ejpe.2015.06.003>
24. Ming Liang, Xue Xin, Weiyu Fan, Hao Wang, Shisong Ren and Jingtao Shi (2017), Effects of polymerized sulfur on rheological properties, morphology and stability of SBS modified asphalt. Construction and Building Materials 150 (2017) 860-871, DOI: <http://dx.doi.org/10.1016/j.conbuildmat.2017.06.069>
25. Naipeng Tang, Weidong Huang, Jianying Hu & Feipeng Xiao (2017), Rheological characterisation of terminal blend rubberised asphalt binder containing polymeric additive and sulphur. Road Materials and Pavement Design, DOI: 10.1080/14680629.2017.1305436
26. Wachira Saowapark, Chanchira Jubsilp & Sarawut Rimdusit (2017), Natural rubber latex-modified asphalts for pavement application, effects of phosphoric acid and sulphur addition. Road Materials and Pavement Design. DOI: 10.1080/14680629.2017.1378117
27. Ashlyn D. Smith, Rhett C. Smith and Andrew G. Tennyson (2020), Polymer cements by copolymerization of waste sulfur, oleic acid, and pozzolan cements. Sustainable Chemistry and Pharmacy 16 (2020) 100249, DOI: <https://doi.org/10.1016/j.scp.2020.100249>
28. <https://www.statista.com/statistics/1031181/sulfur-production-globally-by-country/>