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Orginal Article

Effects of crumb rubber and styrene-butadiene rubber additives on the properties of asphalt binder and the Marshall performance properties of asphalt mixtures

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Abstract: The primary aim of this study is to evaluate the impact of incorporating crumb rubber (CR) and styrene-butadiene rubber (SBR) additives, ranging from 0% to 5% by weight of bitumen, on the performance of a bituminous concrete mixture using the wet process. Laboratory experiments, including the Marshall test, were conducted to establish the optimum bitumen content (OBC) for the hot mix. The study focuses on determining the optimal proportions of CR and SBR to achieve maximum strength. The results show that increasing the proportions of both CR and SBR leads to significant improvements in strength, with the maximum stability recorded at 16.14 kN and a flow of 1.23 mm for a mix containing 5% CR and 4% SBR. The findings further suggest an inverse relationship between CR content and strength, while an increasing SBR content enhances strength. Consequently, the optimal proportions for incorporating CR and SBR additives are identified as 5% and 4%, respectively.

Keywords: Crumb rubber, styrene-butadiene rubber, fly ash, hot mix asphalt, Marshall properties

1. Introduction

There are 1.5 billion tyres produced annually in the world, and after a certain period, these will be considered end-of-life tyres (ELTs). In 2010, about 3.3 million tonnes of used tyres underwent environmentally responsible processing. Based on data regarding the

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planned reuse of tyres, it has been estimated that a substantial amount of approximately 2.7 million tonnes of ELTs would be left as waste [1].

The recycling of automobile tyres and the reclamation of asphalt pavement yield recycled tyre rubber and reclaimed asphalt pavement, respectively. A large amount of waste disposed of in landfills increased the scarcity of landfills and posed a threat to water [2].

Since the late 1960s, crushed tyre rubber has been added to asphalt cement binders as a modifier. This modified binder was first used as a chip seal binder on pavements in Phoenix, Arizona [3].

The CR-modified asphalt mixes made with the wet process are likely better at resisting cracks in pavements compared to conventional and other polymer-modified asphalt (PMA) mixes [4-6]. The air voids and mineral aggregate voids are two key factors affecting the rutting behaviour of wet process CR mixtures [7]. Wet-process CR mixes show lower stiffness and increased susceptibility to moisture damage when compared to PMA mixes [8].

In the context of flexible pavements, the use of CR has emerged as a promising approach to improve the properties of asphalt binders. The CR is more durable and has a more elastic surface than conventional asphalt. When used on highway pavement, CR decreases rutting on new pavement, increases the age and skid resistance of the pavement, and reduces noise levels [9]. The grinding method used for tyre processing significantly affected both the surface area of the tyres and the properties of the asphalt binders [10]. The CR improves the asphalt mixture's water resistance, minimising road damage, with a CR percentage of 25% being optimum [11].

With an increase in the CR content, the modified asphalts' softening point, flash and fire points, and viscosity exhibited a significant increase, while the ductility and penetration experienced a decrease. It may be stated that using CR to modify asphalt binders is a successful and helpful strategy [12]. Styrene-butadiene-styrene (SBS) modified asphalt mixtures are commonly used in road construction due to their remarkable performance. They are also widely applied as wear courses on bridge decks and drainage asphalt pavements [13-15]. The Marshall Mix Design was used to assess the impact of CR in the mixture on volumetric properties, and mixture performance. The addition of fine CR to asphalt mixtures led to the enhancement of various properties, potentially owing to the presence of partial contact between the bitumen and rubber particles, which serve as an elastic aggregate in the mixture. Fine CR greatly improved the overall properties of a given asphalt mix [16]. The research analysed the impact of two CR concentrations (1% and 2%) by weight of the asphalt mix, along with two CR particle sizes (No. 40 and No. 80). The primary objective was to evaluate the potential of CR as an effective enhancer of the quality and strength of asphalt mixtures [17]. A higher CR percentage can improve OBC while decreasing Marshall stability [18]. The Marshall stability and flow properties were improved by using an HMA mixture containing 3% phenolic resin (PF) and 4% SBR by weight of asphalt binder [19]. The study explored the influence of CR on engineering parameters by gradually increasing the CR content in the mixture from 0% to 3%. The results of the analysis revealed that the optimum CR content for the mixture is in the range of 1.5-2% [20]. The study found that asphalt mixtures modified with SBR and polystyrene (PS) exhibit better performance compared to conventional asphalt mixtures. The recommended polymer content for SBR and PS is 5% by weight of asphalt to improve HMA performance [21]. The utilisation of various CR particle sizes and SBS has resulted in an improvement in the cost-effectiveness of asphalt binders [22]. As the CR size increased, the permanent deformation values also increased [23].

Many methods of modifying and upgrading conventional asphalts, including the use of chemicals to improve their properties, have been researched in recent years. Because waste tyres develop quickly and are difficult to dispose of, they are a serious environmental concern that many countries face. Incineration of millions of ELTs is not considered a sustainable or environmentally friendly solution. The CR material for asphalt paving is the sole remaining prospective market for CR. In recent decades, the use of discarded tyres as a material for road construction has become a widespread practice to alleviate environmental burdens.

The primary aim of this study is to assess the impact of CR and SBR on the Marshall properties of HMA mixtures. This research investigates the combined influence of CR and SBR on the Marshall properties of bitumen and asphalt mixtures to achieve the benefits of the CR modifier and enhance bitumen and asphalt. This study's primary objective is to carry out a laboratory investigation into the effects of modified CR and SBR asphalt binders and mixtures.

2. Research methodology

Figure 1 shows the entire process of the research methodology. The materials involved in this study are asphalt binder, fine and coarse aggregates, fly ash (FA), CR, and SBR. The total number of Marshall specimens made for Marshall and flow testing was 90, with 60 for OBC and three for each per cent of CR and SBR. The laboratory-based evaluation of asphalt, CR, aggregates, and mixes was conducted to assess the Marshall properties of asphalt mixtures in accordance with the NHA 1998 guidelines. The study was carried out in Peshawar, KPK, Pakistan.

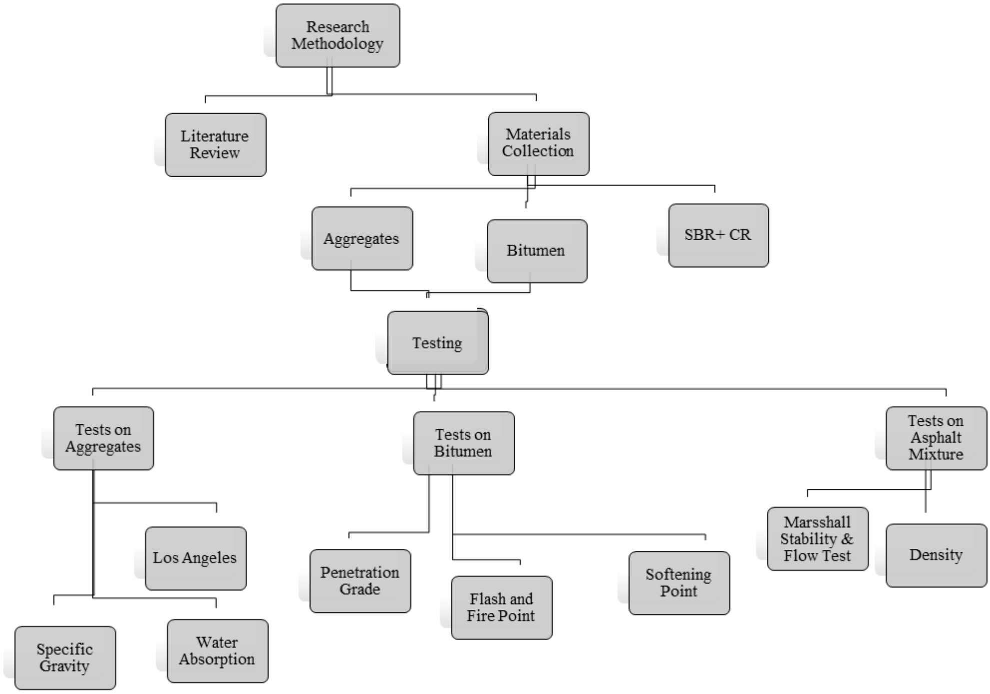

Fig. 1. Research flowchart

2.1 Materials

2.1.1 Asphalt cement

Asphalt is classified as a viscoelastic material, indicating that its physical characteristics are influenced by temperature and loading time. The asphalt is viscous during mixing and compaction, but viscoelastic over its service life. During the manufacturing and service life processes, asphalt's composition allows it to cover and hold aggregate [24]. The bitumen used to make Marshall samples had a penetration grade of 80/100. Prior to use, the grade was double-checked.

2.1.2 Crumb rubber

Crumb rubber (CR) is a recycled rubber material obtained from discarded vehicle tyres. Steel and tyre cable are removed during the recycling process, leaving granular tyre rubber. A granulator or cracker mill is used to further reduce the particle size, sometimes with mechanical help. Particles are sized and categorised according to a variety of factors, including colour (black only or black and white). The CR is a sustainable material since it is made from recycled rubber and does not disintegrate in soil or groundwater. The utilisation of waste rubber from various rubber products has the potential to decrease rubber costs and reduce the volume of rubber waste in the environment. Rubber is used to impart certain properties to several items. The nature of the asphalt-CR mixture may be influenced by some CR properties. The CR (40 mesh crumb) used in this research was obtained from Chakwal, Punjab. It's made from black granules recycled from old tyres. Three different percentages (3, 4, and 5%) of CR per weight of asphalt cement were used to determine the optimum percentage of CR required. The physical characteristics of asphalt rubber are influenced by the overall rubber hydrocarbon content, and the presence of natural rubber content adds to these effects [25].

2.1.3 Styrene butadiene rubber

Styrene-butadiene rubber (SBR) is the world's most frequently used synthetic rubber. Production was established on a massive scale in the United States and Germany during World War II. Styrene and butadiene are polymerised in a solution or emulsion to make them [26]. The SBR polymers are used to increase the binder's adherence. The SBR chemicals were collected from a company near Rawalpindi, Punjab. To determine the optimal percentage of SBR content, three varying weight percentages of SBR (3%, 4%, and 5%) were utilised.

2.1.4 Filler

Fly ash (FA) is a finely divided residue, predominantly composed of spherical, vitreous particles, generated as a by-product during the combustion of pulverised coal in thermal power plants [27]. After incomplete combustion, "fly ash" is a fine powder made from non-combustible coal sources that retains a small amount of carbon [28]. The FA refers to the tiny coal ash particles carried away from the combustion chamber by exhaust gases. The FA, sometimes known as fly ash, is a light brown powder with silt- or clay-sized glassy particles. The chemical and physical composition of FA is highly dependent on multiple factors such as the source of coal, combustion conditions, and the collection method employed at the power plant [29]. The FA was acquired from one of the suppliers in Rawat, Rawalpindi, Punjab. The specific gravity of FA is 2.2. In this study, 4% FA was used as a filler substitute in the mixture as the optimum filler content [30]. The results of the Marshall stability test showed that the optimum filler content was 4% marble dust [5]. The Marshall stability and flow test results showed that the samples prepared with 3% coal bottom ash as filler and an OBC of 4.53% satisfied the NHA requirements for flexible pavement [31]. The results revealed that surkhi, natural sand, stone dust, and cinder all showed a diminishing tendency in developing interlocking properties in asphalt mixtures at internal friction angles of 35°, 33.7°, 32°, and 28.4°, respectively [32].

2.1.5 Aggregate

Aggregate is a fundamental constituent of transportation infrastructure, particularly in the context of road pavement. Aggregate refers to any material that accounts for 90 to 95% of the overall weight of the mixture or 75 to 85% of the volume [33].

An aggregate refers to a composite material composed of solid minerals in the form of large fragments or masses, including coarse aggregates, sand, and aggregate dust. Before incorporating an aggregate into a pavement mixture, it is necessary to conduct a laboratory analysis to determine its properties. When determining a suitable aggregate, grain size, cleanliness, porosity, toughness, surface roughness, strength, viscosity, and composition of asphalt must all be considered. The coarse aggregate used in the preparation of the samples was sourced from a construction company located in Peshawar, Pakistan. The aggregate had undergone degradation by 1 inch, and simultaneous sieve analysis was conducted. Aggregate characteristics were tested to ascertain their properties. The test results met the overall specifications outlined by ASTM.

Table 1 shows the results of laboratory experiments on aggregates with general properties. It was calculated as per ASTM standards. Figure 2 shows the particle size distribution curves for the upper and lower limits, as well as the median aggregate grading adopted in this study.

Table 1. Physical properties of aggregates

Test Name	Test method	Results (%)	Specifications
Los Angeles abrasion	ASTM C131	21	<30%
Soundness (Course)	ASTM C88	8	<8%
Soundness (Fine)	ASTM C88	4.3	<8%
Water absorption	ASTM C127	1.02	<2%
Flakiness	ASTM D4791	5.15	<10%
Elongation	ASTM D4791	3.1	<10%
Fractured particles	ASTM D5821	95	>90%
Uncompacted voids	ASTM C1252	46.2	>45%
Sand equivalent value	ASTM D2419	69	>50%

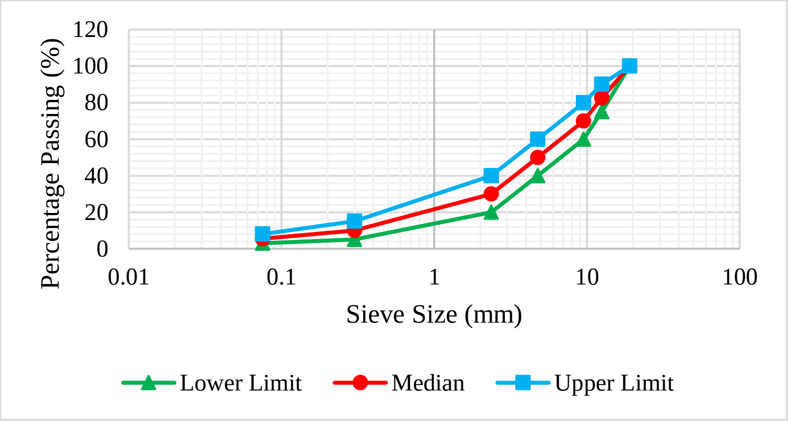

Fig. 2. Particle size distribution curve of aggregate

3. Sample Designations

3.1. Samples prepared with control binder

Table 2 lists the sample designation for the control samples prepared using different binder contents at 4, 4.5, and 5%. The assessment has been carried out based on 3 repetitive samples.

Table 2. Designation, adopted bitumen content, and the number of samples prepared

Designation	Bitumen Content (%)	No. of Samples
Sample C11	4	3
Sample C12	4.5	3
Sample C13	5	3

3.2. Samples prepared with the modified binders

Table 3 shows the designation and details of each sample prepared using different binder types and modifier contents. Essentially, the CR particles used in HMA are finer than 4.75 mm. Most of the particles are between 1.2 and 0.42 mm in diameter. The CR particles can be as small as 0.075 mm in size. It has a specific gravity of 1.10 to 1.20 (depending on how it's made) and must be free of wire, fabric, and other pollutants [34]. In this research, a wet approach is utilized to blend *CR* with the asphalt binder before it is added to the *HMA*. The shredded rubber is directly mixed with the asphalt binder in a mixing apparatus. The blending device blends the CR with the asphalt cement at a speed of 1000 revolutions per minute. The addition of CR was carried out within the first two minutes of the mixing process, followed by continuous mixing for an additional 45 minutes while maintaining the binder temperature at 180°C, in accordance with the experimental procedure [35].

The process used for blending the binder involved the addition of SBR directly to the base asphalt binder at the initiation of the modification process. The laboratory produced the SBR-modified binder by mixing it at a temperature range of 170°C to 190°C for 60 minutes, with a mixing speed of 1000 rpm. This blending process was conducted in accordance with established laboratory procedures [36, 37].

Table 3. Designation, adopted bitumen content and the modifier contents used in the sample preparation

Binder Type	Designation	OBC (%)	CR Content (%)	SBR Content (%)	No. of samples
CR Modified Binders	Sample C21	5	3	----	3
	Sample C22	5	4	----	3
	Sample C23	5	5	----	3
SBR Modified Binder	Sample C31	5	----	3	3
	Sample C32	5	----	4	3
	Sample C33	5	----	5	3
CR+SBR Modified Binder	Sample C41	5	3	3	3
	Sample C42	5	4	4	3
	Sample C43	5	5	5	3

The modified binders incorporating the combination of CR and SBR were also assessed in this experimental study. Prior to the preparation of each CR+SBR-modified binder, the container was filled with a predetermined amount of binder and heated to 170°C. Within the first two minutes, crumbed rubber was added, and the temperature was kept at 190°C for 15 minutes. After the crumbed rubber had been thoroughly mixed, the polymer was added for a 5-minute mix at 1000 rpm.

3.3. Preparation of asphalt mixtures and Marshall stability test

The aggregates were dried for 24 hours at 110±5°C in an oven. The aggregates were then dried to separate them into distinct sieve sizes and recombined in the correct proportion. The weight of the proportion will be precisely 1200 grams. The bitumen should be heated on a hot plate for no more than 30 minutes, and it should always be stirred to avoid local overheating. The temperature should remain constant for at least 10 minutes. The dry blended aggregate was combined with the required amount of asphalt cement that had been prepared. The aggregate was then thoroughly mixed, and the mixture's temperature was kept constant.

To proceed with the compaction process, the loose mixture was poured into the moulds and vigorously stirred with a preheated spatula. Then, each sample was compacted using the Marshall compactor at 75 blows on both sides and allowed to cool for 24 hours at room temperature. Table 4 shows the specifications for asphalt mixing.

Table 4. Specification of asphalt mixing

Description	Unit	Specification
Asphalt cement	mm	80-100
Mixing temperature	℃	170-180 [30]
Compaction temperature	℃	170-180 [30]

After the density measurements were concluded, the specimens were submerged in a thermostatically controlled water bath, completely immersed and heated for 60 minutes at 60 ± 0.5°C.

Following the completion of the heating phase, the test specimen and heads should be promptly removed from the bath. The specimen should be centered in the lower test head and the upper head should be placed on the slides and aligned with the specimen. The complete assembly should then be accurately positioned underneath the plunger on the testing apparatus.

Position the deformation dial gauge appropriately and then calibrate it to zero or take the initial measurement. It is necessary to have previously calibrated the load ring dial gauge to zero.

A constant load rate of 50.8 mm/min ± 0.5% was applied to the specimen until it reached its maximum load, after which it was reduced, and the maximum load gauge reading was taken. The corresponding deformation (flow) gauge reading was then obtained. Prior to testing additional specimens, the heads were wiped clean.

4. Results and discussion

4.1. Properties of asphalt binders

Table 5 demonstrates the physical properties of asphalt and modified asphalt. Increased CR content was found to have a significant impact on both the asphalt's penetration and ductility, resulting in an elevated softening point. All physical properties of the asphalt met the ASTM standards. The introduction of CR to the asphalt strengthened the modified mixture and reduced its flexibility, as evidenced by a higher softening point and lower penetration. Asphalt's ductility was low, implying that it had poor adhesive properties. The reduced ductility value is related to the manual blending procedure used to create modified asphalt, which influences the physical interactions between bitumen and CR [9]. The process of blending plays a crucial role in producing a uniform asphalt mixture, which is essential in accurately determining the properties of the resulting material.

Table 5. Conventional tests results on asphalt

Type of asphalt mixtures	Amount of additives (%)	Penetration at 25 ºC (dmm)	Softening Point (ºC)	Ductility at 25 ºC	Flash Point (ºC)	Fire Point (ºC)	Specific Gravity at 25/25 ºC
Test method		ASTM D5	ASTM D36	ASTM D113	ASTM D92	ASTM D92	ASTM D70
Base binder results	0	91	49	+100	308	345	1.02
CR content (%)	3	78	55	93	326	364	1.043
	4	76	63	85	361	372	1.020
	5	74	67	79	371	382	1.015
SBR content (%)	3	79	54	96	321	361	1.048
	4	77	61	88	355	371	1.025
	5	75	66	81	367	388	1.013
4% SBR + varying CR	3	77	58	90.5	340.5	367.5	1.034
	4	76.5	62	86.5	358	371.5	1.022
	5	75.5	64	83.5	363	376.5	1.02
Specification		80-100	45-52	<100	>225	>250	1.01-1.05

4.2. Optimum bitumen content

To determine the optimum bitumen content (OBC), the asphalt content was varied between 4-5.5% by weight of the asphalt mixture, and three samples were evaluated for each modification. The results of the Marshall Test, which met the requirement specifications, were used to determine the OBC. Table 6 presents the optimal asphalt quantities for various CR and SBR percentages.

Table 6. Optimum asphalt content

Type of asphalt mixture	Control sample	CR			SBR			4% SBR + varying CR		
Amount of additives (%)	0	3	4	5	3	4	5	3	4	5
OBC (%)	5	5			5			5		

4.3. Marshall test results

The results of the Marshall test for all the asphalt concrete mixtures are presented in Table 7. For these Marshall and flow tests, the OBC for each asphalt concrete mixture was determined. Because all the parameter values are within the required criteria, using CR and SBR as additional ingredients in the asphalt mixture produces good results.

Table 7. Comparison of Marshall test results for all asphalt concrete mixtures

Description	Amount of additives (%)	OBC (%)	Marshall stability (kN)	Flow (mm)	VMA (%)	VFB (%)	Marshall Quotient (kN/mm)
Control sample	0	5	6.3	2.7	17.81	74.34	2.3
CR	3	5	5.4	3.5	14.62	74.30	1.6
	4		9.9	2.9	14.13	74.36	3.4
	5		9.2	3.2	14.00	74.70	2.9
SBR	3	5	7.9	2.4	14.98	74.89	3.4
	4		10.3	2.1	13.67	74.93	4.9
	5		12.4	1.8	13.14	73.97	6.9
4% SBR + Varying CR	3	5	10.3	3.1	14.14	74.62	3.3
	4		13.1	2.9	13.9	74.64	4.5
	5		16.1	1.8	13.84	74.82	8.9
Requirements			<5.88	<2-4	<13	65-75	2-5

Based on the Marshall test results, it was observed that the increase in the CR and SBR contents led to an increase in both the Marshall stability and quotient, while causing a decrease in the flow value. The incorporation of CR and SBR in the asphalt mixture resulted in improved strength and quality, as indicated by the higher Marshall Quotient values. The higher values of Marshall Quotient signify greater stiffness and resistance to cracking in the asphalt mixture. As a result, the asphalt mixture resists permanent deformation more effectively. It has also been observed that when there is a high amount of CR and SBR in the mix, flow decreases (Figure 6 and 8). A lower flow number indicates a lack of the asphalt component in the mixture, which causes it to stiffen. It seems as if the optimum asphalt content is decreasing. The CR and SBR were also shown to reduce the quantity of voids filled with bitumen (VFB). The asphalt-filled voids in mineral aggregates are referred to as VFB. As VFB decreases, effective asphalt film thickness decreases, resulting in reduced asphalt mixture durability. The quantity of voids in the mixture (VIM) tends to increase as the CR and SBR percentage rises. The content of the air voids is displayed in VIM. The amount of asphalt in a mixture reduces as the number of air voids increases. A drop in the optimum asphalt content demonstrates this. The combination of CR and SBR in asphalt concrete mixtures can reduce the voids in mineral aggregates (VMA), which can improve the stability and durability of the mixture. However, this reduction in VMA also reduces the space for the asphalt film, which is necessary for a long-lasting asphalt mixture. Therefore, finding the optimal combination of CR and SBR is crucial to achieving both improved stability and durability and a thick asphalt film.

4.3.1. Marshall stability

The addition of finer and harder SBR and CR particles to asphalt cement particles has enhanced the stability of the asphalt mixture. This improvement in stability can be attributed to the presence of these particles in the mixture. The viscosity of the mixture has also increased due to the stiffness of the CR and SBR. In Figure 9, a graphical representation of the correlation between additive percentages and stability is presented. The stability value of a mixture of modified CR and SBR is determined by the Marshall test results. As the contents of the mixture increase, the stability value also increases until it reaches the optimal value when the asphalt content is at its maximum, as illustrated in Figure 3.

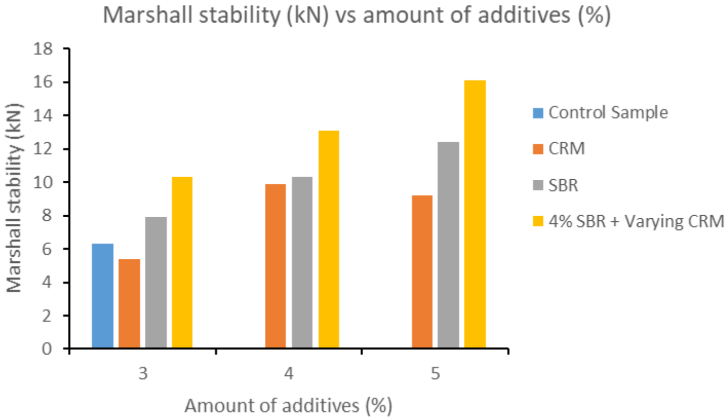

Fig. 3. Stability comparison. Note: CRM = Crumb Rubber Modifier

The stability value obtained is more than 5.88 kN, which meets all AASHTO specifications. The mixture with modified CR and SBR of 4% CR and 5% SBR has the highest stability value, with a stability value of 16.14 kN. The optimum modified SBR and CR contents are 4% and 5%, respectively.

4.3.2. Marshall flow

Flow values of both asphalt mixtures versus bitumen content are shown in Figure 4. Flow decreases as the CR and SBR contents increase. The flow value of the mix with 4% SBR optimum content was 2.9 mm, whereas the flow value of the mix with 5% CR optimum content was only 1.8 mm at the OBC. Both mixtures met AASHTO's requirements.

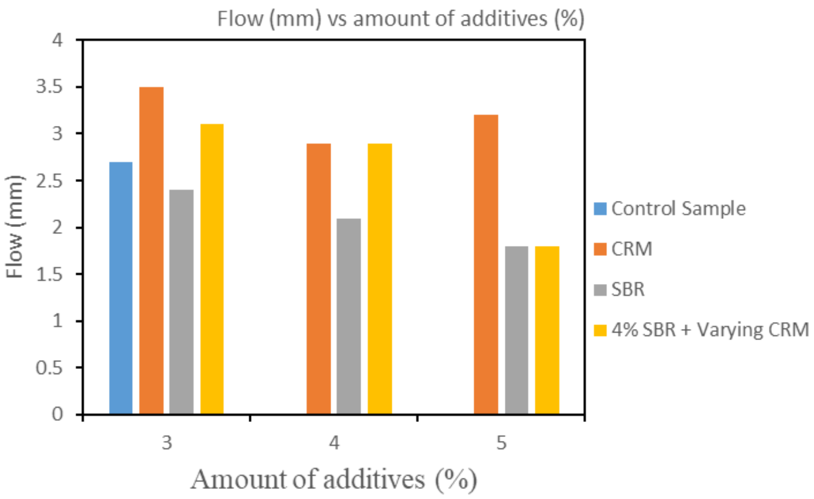

Fig. 4. Flow comparison

4.3.3. Marshall quotient

The Marshall Quotient value obtained is in accordance with AASHTO requirements, which require a minimum of 2-5 kN/mm. The mixture with the lowest Marshall Quotient value of 1.6 kN/mm has a 3% CR content, while the mixture with the highest Marshall Quotient value of 8.9 kN/mm has a 4% SBR and 5% CR. The Marshall Quotient of a mixture containing 4% SBR content is 4.9 kN/mm, while that of a mixture with 5% CR content is 2.9 kN/mm, and a mixture with 4% SBR and 5% CR content is 8.9 kN/mm. However, due to their low stability and high flow, the Marshall Quotient Mixture Modified SBR and CR have a low value, and the coated aggregates become thick and easy to alter, reducing the binding capacity between the aggregates in the mixture when loaded. The reduced binding between aggregates will negatively affect the mixture's stability, leading to a higher flow value as illustrated in Figure 5.

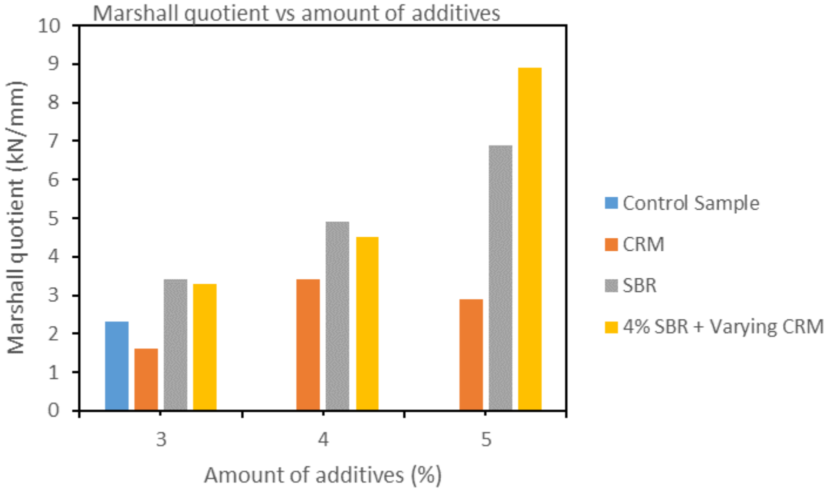

Fig. 5. Marshall Quotient vs amount of additives

4.3.4. Voids in mineral aggregates

The voids in the total mix are plotted against the percentages of additives in Figure 6. The asphalt containing CR, SBR, and FA has fewer voids than the control sample at the OBC. Even though CR, SBR, and FA modified asphalt mixtures have more rigid voids than conventional asphalt mixtures, the smaller voids in the modified mix are due to the improved bonding achieved as a result of the asphalt surface alteration and stiffening effect.

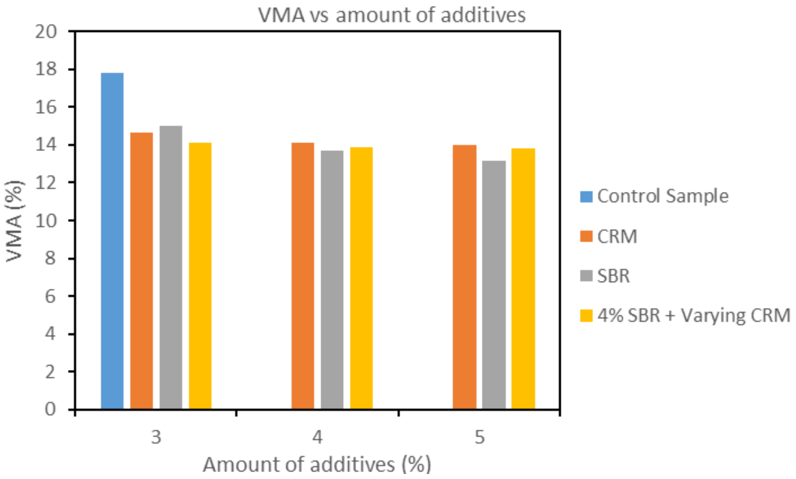

Fig. 6. VMA vs amount of additives

4.3.5. Voids filled with bitumen

Figure 7 depicts the amount of bitumen in the void versus the amount of additives. When the amount of bitumen increases in both mixtures, the number of voids filled with bitumen increases. Furthermore, at the OBC, CR, SBR, and FA modified asphalt mixtures filled a higher percentage of voids with bitumen than conventional asphalt mixtures. It is to be expected that modified asphalt mixtures have a lower void content overall.

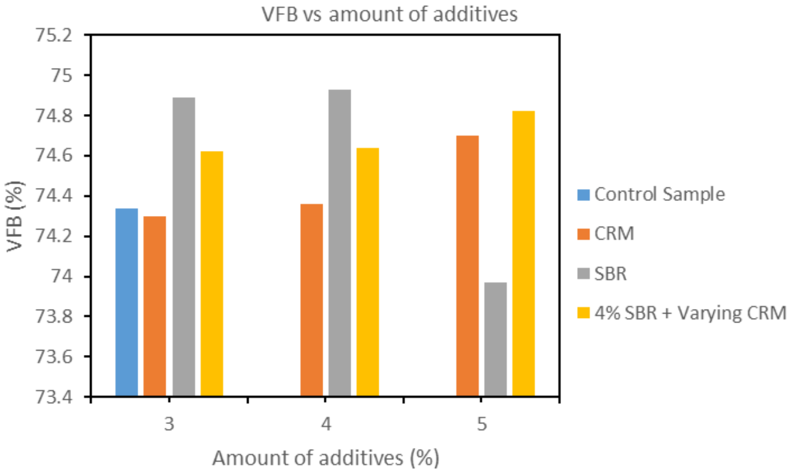

Fig. 7. VFB vs amount of additives

The optimum amount of CR and SBR content is the amount at which the specimen has the highest Marshall stability and flow rating. Figures 4 and 6 show that the optimum CR content is 5% and the SBR content is 4%. Based on the presented figures, an increase in the content of CR and SBR in asphalt mixtures results in a decrease in the values of Marshall Stability and Marshall Flow.

5. Conclusions

It was determined that the modified mix had better Marshall Properties, as listed below, based on the behaviour of SBR and CR modified bitumen content.

- Increasing CR and SBR to 5% and 4% enhances Marshall Stability. However, beyond 5%, the flow improves but stability decreases, suggesting reduced resistance to heavy wheel loads.
- Increasing the proportions of both CR and SBR results in notable improvements in strength, with optimal proportions determined to be 5% CR and 4% SBR.
- The study provides valuable insights into the inverse relationship between CR content and strength, and the positive impact of increasing SBR content on strength, offering practical recommendations for incorporating CR and SBR additives in bituminous concrete.

It is suggested that additional studies into the problem be conducted and that more trial sections be established. To improve the properties of bitumen mix, polythene bags and other binders can be used.

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