https://doi.org/10.22581/muet1982.2303.07

2023, 42(3) 59-66

Performance characterization of crumb rubber modified bitumen using pyrolyzed waste tyre treated bitumen

Asim Shaikh *, Naeem Aziz Memon, Aneel Kumar, Ghulam Yasin Shaikh

Department of Civil Engineering, Mehran University of Engineering and Technology Jamshoro

* Corresponding author: Asim Shaikh, Email: engr.asimshaikh@gmail.com

Received: 24 May 2023, Accepted: 26 June 2023, Published: 01 July 2023

KEYWORDS

Crumb Rubber

Bitumen

Pyrolysis Process

Polymer

Thermogravimetric Analysis Test

ABSTRACT

Exponential deterioration in pavement caused by heavy loads, temperature variations, and heavy rainfall had led to pavement failure. To overcome those failures, bitumen properties needed to be enhanced in an economical and sustainable way. Researchers have used various modifiers to enhance the properties of virgin bitumen, but the end product still does not seem to be accepted by the asphalt industry at the required level. On the other hand, the construction cost has also increased due to the addition of modifiers, which have varying performance characteristics. To address such issues, waste materials such as crumb rubber obtained from waste tyres are used to enhance the properties of bitumen. The incorporation of crumb rubber has enhanced the properties of bitumen and has been well proven for decades. However, the product is still not widely accepted due to limitations such as consistency and uniformity during and after the production stages. In this research, crumb rubber was converted into pyrolyzed oil using a pyrolysis protocol and it was observed through the values of penetration and softening point test shows that optimum 2% pyrolyzed oil can be treated with bitumen to increase workability and flowability at low temperatures, such that additional crumb rubber in crumb form can be added homogenously. The results showed that an optimum quantity of 20% crumb rubber by mass of bitumen can be blended with pyrolyzed modified bitumen that is 5% crumb rubber more compared to untreated modified crumb rubber bitumen, which simultaneously increases the physical and mechanical properties of the mix. It enhances the softening point, viscosity, and storage stability while decreasing the penetration value.

1. Introduction

Transportation infrastructure plays an essential role in the socio-economic development of a country. It may be by land, air, or water transportation; each has its own significance. The most widely used mode of transportation is land transportation, which includes the railway network and the road network. The most widely used mode of land

© Mehran University of Engineering and Technology 2023

transportation globally is roads because of their cheap and safe access. Road networks provide sources of communication, trade, education, access to employment, and health services, which act against poverty and illiteracy rates that directly affect the gross domestic product (GDP) of a country, and due to these, the economic and social conditions of the country are improved. Population growth has led to an increase in demands for heavy loads and traffic volume, which has degraded road quality. The preservation of this vast infrastructure needs appropriate and cost-effective design techniques and materials so that the life span of roads and road quality may be improved.

95% of roads in the world are made of flexible pavement in which bitumen acts as a binding agent. Bitumen has worldwide applicability in pavement construction; its performance is required and needs to be enhanced including tensile strength, resistance to ageing, chemical and thermal stability, mechanical strength, fatigue life, moisture susceptibility, temperature susceptibility, rutting resistance, viscosity, stiffness, storage stability and many more [1]. Bitumen is a viscoelastic material used as a binder in flexible pavement. The viscoelastic property sets bitumen at room temperature in solid form but at low temperatures it becomes brittle while at high temperatures it becomes flowable. To resist thermal cracking bitumen should be elastic and soft at low temperature while stiff at high temperature to resist rutting [2]. Most virgin bitumen doesn't have the required properties which causes failures. The failures are caused by the high stresses and temperature variation, so modifiers are used to increase the rheological properties of bitumen by blending it with polymers like plastic and rubber [3]. Researchers have used many modifiers to enhance asphalt properties but alternately they increase the cost of pavement. To identify a sustainable solution, waste material such as crumb rubber should be used that is available in ample quantity and would be a solution to non-biodegradable waste.

Crumb rubber in asphalt pavement transfers the property of elasticity. According to research, the modification of bitumen by crumb rubber at high temperature increases the complex modulus of the binder while reducing the phase angle which has an effect on resistance to rutting. The advantage of crumb rubber modified asphalt pavement is that it is durable, resistance to permanent cracking and thermal deformation [4]. The incorporation of CR reclaimed from waste tyres has resulted in enhanced properties in conventional bitumen. The engineering benefits from such a modification of bitumen are very well proven and have been recognized around the world for decades. However, the product is still not widely accepted and used in the pavement industry like other polymer modified bitumen products. The main reasons for this are some limitations, such as consistency and uniformity during and after the production stages © Mehran University of Engineering and Technology 2023

respectively, with a breakthrough yet to be achieved in the form of a stable modified bitumen without compromising the properties of the product. In CRMB (crumb rubber modified bitumen) the crumb rubber absorbs partially the lighter components in bitumen while remaining swelled up to nine times as per the volume compatibility problem. Due to the nonpolar chemical nature and stability of the blend it becomes heterogeneous (phase separation occurs) [5] [6]. ASTM 2009 defines the that the maximum quantity of crumb rubber as a modifier is 15% of the weight of bitumen. Many researchers have concluded that 4% of crumb rubber in bitumen has no significant effect on the mechanical and performance properties of the binder and that more than 20% is unsuitable [7]. The direct blending of crumb rubber into bitumen produces secondary environmental pollution [8]. To stabilize in a sustainable way, bitumen is pre-treated with pyrolyzed oil by which a chemical reaction occurs and the bond in bitumen breaks which accelerates the dissolution of crumb rubber in bitumen enhancing mechanical properties.

2. Material Properties and Experimental Program

2.1 Material Properties

The properties of the crumb rubber gained from truck tyres are shown in Table 1. Truck tyres were sliced into chunks and then the steel wires were removed, which were then physically crushed into a crumb that could pass through a #40 sieve. The crumb rubber's density in relation to water was determined to be 1.14, which is indicated by its specific gravity. The temperature at which crumb rubber begins to catch fire when exposed to a flame was determined to be its flash point, which is 380°C and the thermal breakdown temperature of crumb rubber was 201°C.

Table 1

Crumb rubber properties

Properties	Crumb Rubber	
Size	Crumb form (#40 passing)	
Туре	Truck tire rubber	
Specific gravity	1.14	
Flash Point	380°C	
Thermal Decomposition	201°C	

The bitumen of 60/70 grade was used because it is costeffective, reliable and capable of increasing performance through its penetration index and softening point values which are idle for hot and moderate temperatures. The properties of bitumen of 60/70 grade are shown in Table 2. Table 2

Properties of 60/70 grade bitumen

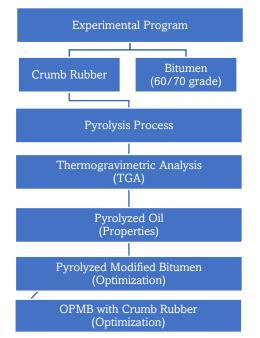
Properties	Results	Range	ASTM Codes
Specific Gravity@25°C	1.018	1.01-1.06	D-70
Softening Point	49°C	48-56°C	D-36
Ductility@25°C	100 +	100 min	D-13
Flash Point	336°C	240°C	D-92
Penetration@25°C	69	60-70	D-5
Fire Point	380°C	240°C	D-92

2.2 Experimental Program

Following are the steps to conduct this experimental program:

- a) The first step was to produce oil from crumb rubber. The pyrolysis method was used to thermally convert crumb rubber into oil. The optimum rate of heating was found through thermogravimetric analysis.
- b) The second step was to add pyrolyzed oil to bitumen at varying percentages and calculate the optimum content based on penetration and softening point values.
- c) The third step was to add optimized pyrolyzed modified bitumen with different percentages of crumb rubber to calculate the optimum crumb rubber content based on the penetration, viscosity, softening point and storage stability tests.

Following is schematic diagram of the experimental program plan.



3. Results and Discussion

In this research, bitumen is treated with pyrolyzed oil and then modified with crumb rubber. Pyrolyzed oil is obtained from crumb rubber by the pyrolysis method. Pyrolysis is a method to thermally breakdown the material and convert it into fuel in the absence of oxygen. It is an eco-friendly process with no secondary environmental pollution [9]. Before starting the pyrolysis process, the thermal decomposition of material is to be found by thermogravimetric analysis, through which the rate of heating and optimum temperature could be determined.

3.1 Thermogravimetric Analysis Test (TGA)

In Fig. 1, it shows the TGA device was connected to a computer and a cylinder containing nitrogen gas, which was used to remove oxygen and create an inert environment because the test must be done in the absence of oxygen. A TGA test of crumb rubber was conducted to determine the range of temperature with respect to time. The test was conducted at different heating rates (50, 100, 150, and 200 °C/min). The results in Fig. 2 showed a symmetrical change with increasing rate of heating, and it could be concluded that rate of heating has no effect on material (crumb rubber), so 50°C/min was decided. The graph pointed out that the crumb rubber starts decomposing at 200°C, while the removal of volatiles starts at 250°C to 550°C, after which the degradation starts. The pyrolyzed oil is produced during the removal of volatiles from the material, and after removal, the carbon black is left as residue in the pyrolysis reactor.



Fig. 1. Thermogravimetric analysis test (TGA) setup

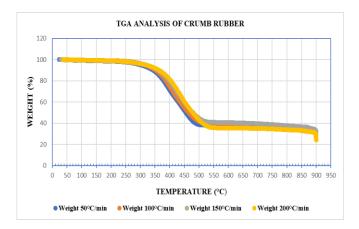


Fig. 2. TGA test analysis of crumb rubber

3.2 Production of Pyrolyzed Oil

The laboratory pyrolysis apparatus was designed as shown in Fig. 3. The cylindrical-shaped container (feeding chamber) is inserted into the muffle furnace and heated at 550° C.

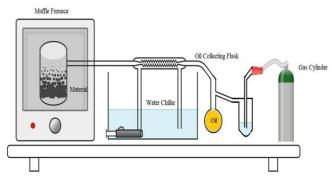


Fig. 3. Laboratory pyrolysis apparatus

The main chamber is connected to the water condenser at 5°C temperature. Oil produced from the material is collected in an oil collecting vessel, while gas is collected in a gas cylinder. Between the oil collecting flask and the gas cylinder, there is an aqueous condensate beaker attached to the oil collecting flask to retain the maximum amount of gas to convert into vapors in the oil flask while the rest of the gas flows through the aqueous medium into the gas cylinder. The properties of pyrolyzed oil are shown in Table 4.

Table 3

Properties of pyrolyzed crumb rubber oil

Properties	Pyrolyzed Crumb Rubber (Oil)		
рН	3.4		
Specific Gravity	0.81		
Flash Point	84°C		
Fire Point	90°C		
Condensing Temperature	19°C		
Viscosity	4.2		

3.3 Optimization of Pyrolyzed Modified Bitumen

© Mehran University of Engineering and Technology 2023

The pyrolyzed crumb rubber oil was blended with 60/70 grade bitumen at 1%, 2%, 3%, 4%, and 5%. The optimization of percentages was based on penetration and softening point graphs. The pyrolyzed oil was blended at 100 rpm for 5 minutes with bitumen at 80°C because the flash point of pyrolyzed oil is 84°C and fire is 90°C.

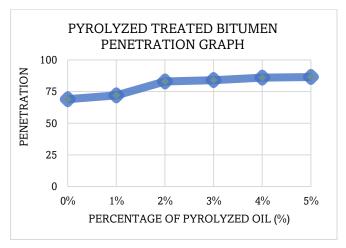


Fig. 4. Pyrolyzed oil treated bitumen penetration graph

In the penetration graph, it could be observed that the maximum change in variation occurred at 2% of pyrolyzed modified bitumen, while after that there was a slight variation in the penetration value while the same trend went in the softening point. According to the results, the optimization of pyrolyzed crumb rubber modified bitumen is at 2% of pyrolyzed oil. Adding an additional quantity of pyrolyzed oil has a slight variation, but it was observed that excess oil floats on the surface of the container. The trend describes the variation: as the percentage of oil is added to bitumen, the penetration value increases, leading to thinner bitumen, while the softening point reduces, which makes asphalt flowable at low temperatures. It can be used extensively for bitumen seals as a spray to retard crack propagation, increase adhesion in aggregates, and as a strain-alleviating membrane (SAM).

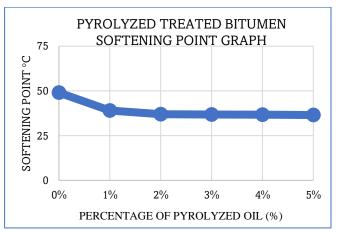


Fig. 5. Pyrolyzed oil treated bitumen softening point graph

3.4 Optimized Pyrolyzed Modified Bitumen with Crumb Rubber:

Crumb rubber modified bitumen is produced in various parts of the world by many different agencies that depend on the production, usage, processing, and type of crumb rubber. Several specifications have been published and implemented for modified crumb rubber. In this research, 2% optimized pyrolyzed bitumen was blended with different proportions of crumb rubber to determine the optimum content of crumb rubber in pyrolyzed bitumen. The optimized modified crumb rubber limits were determined from ASTM D6114 specifications, which are mentioned in Table 5, and material Type II grade is used.

Table 4

ASTM D 6114 specifications,2002

Properties	Type I	Type II	Type III
Apparent Viscosity	1500-	1500-	1500-
	5000	5000	5000
Penetration	25-75	25-75	25-75
Softening Point	<u>></u> 57.2	<u>></u> 54.4	<u>></u> 51.7
Flash point	<u>></u> 232.2	<u>></u> 232.2	<u>></u> 232.2
Specific Gravity	1.15+-	1.15+-	1.15+-
	0.05	0.05	0.05

A comparison is made between crumb rubber modified bitumen and crumb rubber modified pyrolyzed bitumen. Crumb rubber modified bitumen was prepared by a wet process. Crumb rubber at 0, 10, 12.5, 15, 17.5, 20, and 22.5% percentages was added to virgin bitumen and pyrolyzed bitumen individually to determine optimum content based on ASTM D6114 standards. The stability of the modified mix was calculated by analyzing the laboratory tests (penetration, softening point, viscosity, and storage stability). It was observed in the penetration test, as shown in Fig. 6.

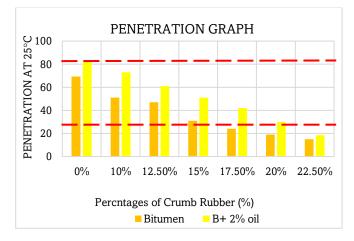


Fig. 6. Penetration of CRMB vs CRPMB

The addition of crumb rubber to virgin and pyrolyzed bitumen decreases the penetration value, making © Mehran University of Engineering and Technology 2023 bitumen hard and brittle while simultaneously increasing shear resistance. In virgin bitumen, the maximum amount of 15% crumb rubber could be used as a modifier, but in pyrolyzed bitumen mixes, the percentage increases to 20%. The pyrolyzed bitumen mix could dissolve crumb rubber more than virgin bitumen due to the pyrolyzed oil treated bitumen, which supports the addition of more crumb rubber to the mix.

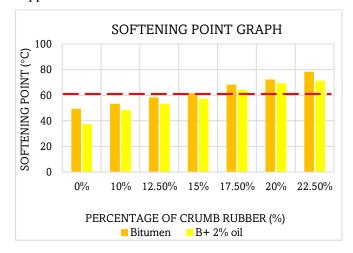


Fig. 7. Softening point of CRMB vs CRPMB

The softening point test is conducted to assess the binder's stability after hot storage. The increase in softening point improves the elasticity of the binder which controls bleeding at high temperatures and resists cracking at low temperatures. In Fig. 7, it can be observed that by adding crumb rubber to virgin bitumen and pyrolyzed bitumen, there is a gradual increase in softening point value. According to ASTM D6144 specifications, the softening point of modified mixes should be greater than or equal to 55°C. The softening point of modified crumb rubber mix was found to be greater than that of pyrolyzed modified crumb rubber mix due to the disbanding of the bond of bitumen.

Viscosity controls the softening and bleeding of the binder, and it was found that the addition of crumb rubber increased the viscosity. The increase in content of crumb rubber in asphalt leads to good fatigue resistance and enhances performance at high and low temperature that alternatively increases the viscosity of the modified mix while resulting in poor workability.

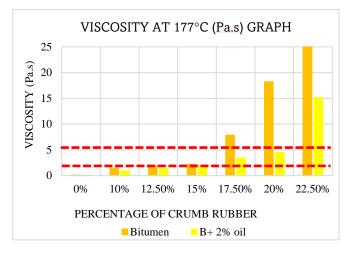


Fig. 8. Viscosity at 177°C of CRMB vs CRPMB

Therefore, pyrolyzed oil is used to enhance the content of crumb rubber and reduce its viscosity by breaking the long chain molecules of rubber by depolymerization and generating free radicals that make crumb rubber soluble in asphalt mix. The increase in viscosity enhances the stability of the pavement by reducing the rate of stripping. The workability of modified bitumen should be considered when viscosity is within limits. In this research, viscosity was calculated using a Brook field viscometer and the limits for modified crumb rubber bitumen mix are 1.5 Pa.s to 5 Pa.s. In Fig. 8, the addition of crumb rubber to bitumen increases the viscosity. It was found that a maximum of 15% of crumb rubber can be added to virgin bitumen and 20% to pyrolyzed bitumen which is due to the pyrolyzed oil that dissolves the crumb rubber in bitumen.

The resultant physical changes in the properties of the modified bitumen mix can be investigated by a storage stability test. The storage stability test was conducted in accordance with ASTM D5976 / ASTM D36 specifications by considering the objective of understanding the phase separation of polymer dispersion in bitumen. If the difference in softening points of the top and bottom-third sections after 6 hours of prolonged storage at elevated temperatures is less than 2.5°C than the modifier is stable. According to the laboratory test results of penetration value, softening point and viscosity it was found that the optimum content of crumb rubber in bitumen was 15% while in pyrolyzed bitumen it was 20%. In CRMB, certain limitations were observed that by adding crumb rubber more than 15% it resulted in uneven dispersion of the particles in the bitumen mix and the mixture resulted in a heterogeneous mix. In the pyrolyzed modified bitumen mix, it was observed that a uniform layer of mix was formed due to the chemical reaction of pyrolyzed oil with bitumen that accumulated the maximum quantity of crumb rubber and resulted in a homogenous mix.

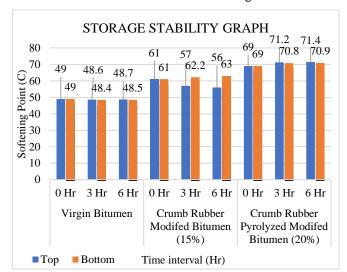


Fig. 9. Storage stability of CRMB vs CRPMB

In Fig. 9, It could be observed that crumb rubber pyrolyzed modified bitumen showed good storage stability as compared to crumb rubber modified bitumen. The difference between the top and bottom values of the softening point showed the nonhomogeneity of the mix in crumb rubber modified bitumen.

Increasing the content of crumb rubber in asphalt increases the service and performance of the road but leads to high viscosity, insufficient thermal storage stability, poor workability and a reduction in the water stability of CRMB **[10]**, while pyrolyzed oil in bitumen is used as a stabilizer due to Sulphur compounds. The pyrolyzed oil in CRPMB decomposes crumb rubber at high temperatures and forms cross links with polymer molecular chains to produce active groups to gain stability. The amount of pyrolyzed oil in CRPMB has limitations because excessive pyrolyzed oil causes severe reactions in asphalt by over cross-linking polymer molecules that form a non-homogenous mix, resulting in instability and weak storage stability.

4. Conclusion

In this research, an experimental study was conducted to evaluate the performance of bitumen treated with pyrolyzed oil obtained from waste tyre rubber. The study aims to assess the effectiveness of this treatment approach in enhancing the properties of asphalt binder. Adding pyrolyzed oil to bitumen breaks the chemical bond because of an acidic reaction. The pyrolyzed bitumen due to the breaking of chemical bonds, allows homogenous modification with crumb rubber without secondary reaction through the mechano-chemical method. The optimum pyrolyzed crumb rubber oil was 2% bitumen while an additional 20% of virgin crumb rubber could be added.

The conclusions obtained are as follows:

- The maximum amount of 15% CR can be blended in bitumen but as compared to pyrolyzed modified bitumen 20% CR can be blended which results in an additional 5% increase in quantity of CR. The pretreatment of bitumen with pyrolyzed oil chemically made voids between the molecules of bitumen due to which additional amounts of crumb rubber could be blended.
- In modified mixes, high temperature is required for blending but in pyrolyzed mixes the addition of CR can be added at a low temperature due to the acidic reaction that occurred during the pre-treatment of bitumen by blending pyrolyzed oil which weakens the chemical bond and reduced the viscosity of bitumen which facilitates the blending at low temperature.
- The CRP.MB resulted in enhanced values of increased softening point and viscosity while decreasing penetration value as compared to the CRMB which simultaneously improved elastic recovery, tensile strength and resistance to ageing. A decrease in penetration value showed that bitumen is hard and a strong binder while an increase in softening point values enhanced the resistance to high temperatures and deterioration in cracks.
- The increase in viscosity of CRP.MB showed resistance to deformation. The addition of CRP.MB was found to control softening and bleeding of the binder at high temperatures and cracking at low temperatures.
- The viscous behaviour of the CRP.MB blends directly affected the penetration values as compared to base bitumen and CRMB. The softer base binders were found to be more capable with CRP.MB by accommodating an increased CR concentration.
- In the storage stability test the difference in the top and bottom values of the softening point were minimal which indicated that CRP.MB has good storage stability. This is attributed to reducing interparticle distance, which prevents the movement of the CR particles. This results in the restriction of excessive settling or floating of CR particles, irrespective of the different densities of the two phases.

Environmental benefits

© Mehran University of Engineering and Technology 2023

- Non-biodegradable waste (CR) is utilized to enhance the properties of the mix that contribute to reduce solid waste pollution.
- A total of 23.3% of crumb rubber is utilized in CRP.MB
- It is an eco-friendly process. In wet and dry processes toxic fumes are generated during blending and require high temperatures which are reduced because this process is conducted at a low temperature.

Amount of energy saved

- The pyrolyzed mix required less effort and a low temperature.
- 20% of the quantity of bitumen is saved which reduces the cost of pavement.

5. References

- [1] S. Sharma, S. Sharma, and N. UPAdHYAY, "Modifier based enhancement in physical and chemical properties of bitumen", 2019.
- [2] D. Lesueur, "The colloidal structure of bitumen: Consequences on the rheology and on the mechanisms of bitumen modification", Advances in colloid and interface science, vol. 145, no. 1–2, pp. 42–82, 2009.
- [3] A. R. Prasad and N. J. Sowmya, "Bituminous modification with waste plastic and crumb rubber", IOSR Journal of Mechanical and Civil Engineering, vol. 12, no. 3, pp. 108–115, 2015.
- [4] S. K. Palit, K. S. Reddy, and B. B. Pandey, "Laboratory evaluation of crumb rubber modified asphalt mixes", Journal of materials in civil engineering, vol. 16, no. 1, pp. 45–53, 2004.
- [5] F. J. Navarro, P. Partal, F. Martinez-Boza, and C. Gallegos, "Thermo-rheological behaviour and storage stability of ground tire rubbermodified bitumens", Fuel, vol. 83, no. 14–15, pp. 2041–2049, 2004.
- [6] M. Porto, P. Caputo, V. Loise, S. Eskandarsefat, B. Teltayev, and C. Oliviero Rossi, "Bitumen and bitumen modification: A review on latest advances", Applied Sciences, vol. 9, no. 4, p. 742, 2019.
- [7] G. Polacco, S. Filippi, F. Merusi, and G. Stastna, "A review of the fundamentals of polymermodified asphalts: Asphalt/polymer interactions

and principles of compatibility", Advances in colloid and interface science, vol. 224, pp. 72–112, 2015.

- [8] T. Ling, Y. Lu, Z. Zhang, C. Li, and M. Oeser, "Value-added application of waste rubber and waste plastic in asphalt binder as a multifunctional additive", Materials, vol. 12, no. 8, p. 1280, 2019.
- [9] R. Dong, M. Zhao, W. Xia, X. Yi, P. Dai, and N. Tang, "Chemical and microscopic investigation of co-pyrolysis of crumb tire rubber with waste cooking oil at mild temperature", Waste management, vol. 79, pp. 516–525, 2018.
- [10] K. Duan, C. Wang, J. Liu, L. Song, Q. Chen, and Y. Chen, "Research progress and performance evaluation of crumb-rubber-modified asphalts and their mixtures", Construction and Building Materials, vol. 361, p. 129687, 2022.
- [11] ASTM, "Standard specification for asphalt-rubber binder", 2009.
- [12] ASTM, "Standard specification for asphaltrubber binder", 2002. D6114 Standard Specification for Asphalt-Rubber Binder, Vol. 4.03, Road and Paving Materials, Vehicle Pavement Systems, Annual Book of ASTM Standards, West Conshohocken, PA.
- [13] G. P. Ganapathy and T. C. Haupt, "Engineering properties of SBS and crumb-rubber modified bitumen–a design of experiment approach", Journal of Engineering, Design and Technology, 2023.
- [14] G. Liu et al., "Emission of volatile organic compounds in crumb rubber modified bitumen and its inhibition by using montmorillonite nanoclay", Polymers, vol. 15, no. 6, p. 1513, 2023.
- [15] S. O. Olopade, B. Kehinde, O. Olawale, and A. A. Olarewaju, "Laboratory study of polymer-modified bitumen in road construction: a review on crumb rubber application", 2023.
- [16] O. Šernas, A. Vaitkus, and J. Škulteckė, "Performance of crumb rubber bitumen and asphalt modified in the wet process alone and in combination with SBS polymer", Road Materials and Pavement Design, vol. 24, no. sup1, pp. 107–123, 2023.

- [17] H. Wang, G. D. Airey, Z. Leng, and G. Lu, "Optimisation of the preparation procedure of crumb rubber modified bitumen with wax-based additives", Road Materials and Pavement Design, pp. 1–12, 2023.
- [18] X. Zhang, X. Wang, C. Wan, B. Yang, Z. Tang, and W. Li, "Performance evaluation of asphalt binder and mixture modified by pre-treated crumb rubber", Construction and Building Materials, vol. 362, p. 129777, 2023.