

Combined Use of Natural Rubber, Biomass and Plastic Wastes in Bitumen Modification and Flexible Pavement Construction



Swapan Kumer Ray, Riyadh Hossen Bhuiyan, Muhammad Saiful Islam, Md. Jaynal Abedin, Zahidul Islam, and Rashed Hasan

Abstract A novel bitumen modification process has been developed using lower grade natural rubber-ribbed smoked sheet (NR-RSS) and soda lignin (SL) isolated from waste green coconut fiber. At first, natural rubber along with rubber additives was mixed in toluene and then soda lignin was added. Obtained mixture was characterized by rotational viscometer, Fourier transform infrared spectrometer, universal testing machine, simultaneous thermal analyzer, etc. The mixture was then blended with 60–70 penetration grade bitumen (B). Physical, spectral, microscopic and thermal properties of modified bitumen (MB) were evaluated. It was shown that the prepared natural rubber-lignin mixture significantly improved the physical properties of bitumen as required for flexible pavement construction in tropical countries. Marshal stability was improved by 25% in comparison to 60–70 penetration grade bitumen. Based on the result obtained, an experimental pavement was constructed using modified bitumen and waste polyethylene-coated aggregates (Pav-MB-PCA). For comparison, another pavement of similar size was constructed with 60–70 penetration grade bitumen and non-coated aggregates (Pav-B-NCA). More than two years observation showed better performances of the pavement made with modified bitumen and waste polyethylene coated aggregates. All findings have been discussed in detail.

Keywords Bitumen · Natural rubber ribbed smoked sheet · Waste green coconut fibre · Waste polyethylene · Marshal stability · Flexible pavement

1 Introduction

Most of the tropical countries have already faced the impact of climate change on the flexible pavement. A significant variation in the daily and seasonal temperature and prolonged rainfall and thus water clogging induces early development of distress condition of bituminous pavement. Adaptive maintenances, such as construction of

S. K. Ray (✉) · R. H. Bhuiyan · M. S. Islam · Md. J. Abedin · Z. Islam · R. Hasan
Fibre and Polymer Research Division, Bangladesh Council of Scientific and Industrial Research,
Dhaka 1205, Bangladesh

permeable pavements, use of modified binders and improved routine maintenances have been recommended by several reports [1]. The incorporation of different polymers and rubbers into bitumen can improve some properties, e.g. improved flexibility, elasticity at low temperature, thermal and crack resistance, moisture resistance, etc. and thus increased service life, etc. [2, 3]. Typically, polymer or rubber modified bitumen are more viscous compared to unmodified bitumen and improve adhesive bonding among the aggregates. Natural rubber could be introduced into bitumen by direct fusion or in presence of suitable solvent(s). But the dispersion capability of rubber is poor in bitumen and thus special measures need to be taken obtaining homogeneous rubber modified bitumen [2]. Some investigations were also carried out on the use of lignin, a highly complex phenolic biopolymer, to prevent bitumen oxidation or hardening [4, 5]. Lignin is a wonderful aromatic biopolymer due to its abundance and relatively low cost [6]. It shows the resistance to oxidation, resistance to water, absorption of ultraviolet radiation, etc. The effect of lignin on the physico-mechanical properties and thermo-oxidative degradation resistance of natural rubber has been investigated [7, 8]. The waste thermoplastics, e.g. polyethylene, polypropylene, etc. could be used in flexible pavement construction by adding them at a certain percentage with aggregates and reported it as a good strategy for utilization of these wastes [9]. So, proper management and utilization of waste thermoplastics in asphalt pavement construction can provide an important outlet for such materials to keep the environment green. The aim of this research work was to examine the properties of natural rubber-lignin composite and its application in bitumen modification. Another objective was to evaluate the moisture and heat resistant properties of flexible pavement constructed with modified bitumen and waste thermoplastic coated aggregates. The overall work might be considered as a solution of proper polymeric waste management and their effective utilization in sustainable pavement construction.

2 Materials and Methods

2.1 Materials

60–70 penetration grade bitumen (B) was used to prepare modified bitumen (MB). Natural rubber ribbed smoked sheet (NR-RSS), waste green coconut fibre, waste high density polyethylene (HDPE) shopping bags, standard aggregates, e.g. granite stone of different sizes and sand were used in this work. Other chemicals used in the experiment were of reagent grade.

Table 1 Preparation of bitumen modifier (BM)

Natural rubber (g)	Toluene (ml)	Rubber additives (g)	Soda lignin (g)	Temperature (°C)	Stirring speed (rpm)	Time (h)	Name of sample
125	1250	0	0	25	1800–2000	4	BM-1
		15	0				BM-2
		15	12.5				BM-3

2.2 Preparation of Soda Lignin

The dried coconut fiber was pre-treated with dilute sulfuric acid and then digested with 3% sodium hydroxide solution at 140 °C for 2 h. After cooling, the slurry was filtered and black liquor was collected. pH of the black liquor was reduced to 2.5 using dilute sulfuric acid and precipitated lignin was collected, purified and vacuum dried (yield 35.32%). Analytically milled soda lignin (SL) powder (particle sizes <2 µm; ash content <0.7%) was used in this work.

2.3 Preparation of Natural Rubber-Lignin Mixture for Bitumen Modification

Natural rubber-lignin mixture, e.g. bitumen modifier sample (BM-3) was prepared following the procedure given in Table 1. Another two samples, BM-1 and BM-2 were prepared for comparison with BM-3. BM-1, BM-2 and BM-3 were cured at 135 °C for 30 min and represented as BM-4, BM-5 and BM-6, respectively.

2.4 Modification of Bitumen and Specification Properties

Modification of bitumen was done by blending BM-3 with 60–70 penetration grade bitumen (B) (5 w/w % natural rubber) at 135 °C under overhead stirring at 2000 rpm for 1 h. Solvent, toluene was collected for reuse. Modified bitumen (MB) was then cooled at room temperature for 48 h and specification properties were determined by ASTM methods. Viscosity of samples, BM-1, BM-2, BM-3, B and MB samples were evaluated by rotational rheometer. Penetration index was calculated using Pfeiffer and Van Doormaal equation [10].

Table 2 Properties of BM-4, BM-5, BM-6, B and MB sample

Properties of sample	Method
Tensile strength and percent elongation	ASTM D 882-02
Curing study	FT-IR spectrometer coupled with diamond ATR
Thermal stability	Simultaneous thermal analyzer
Surface morphology	Scanning electron microscope
Marshal stability	ASTM D1559
Static emersion test	[10]

Table 3 Procedure for the construction of flexible pavements

Granite stones	Plastic used (%)	Temperature (°C) and time (min) for heating	Bitumen used	Bitumen content (%)	Volume of pavement (m ³)	Pavement acronym
Mixture of aggregates for base course	0	130–180 and 30	B	5	9.75 × 9.14 × 0.06	Pav-B-NCA
	2.5		MB			Pav-MB-PCA

2.5 Analysis of Prepared Samples and Construction of Experimental Pavement

2.5.1 Analysis of Prepared Samples

BM-4, BM-5, BM-6, 60–70 penetration grade bitumen (B) and modified bitumen (MB) samples were further analyzed by different methods and presented in Table 2.

2.5.2 Construction of Experimental Pavement

Asphalt base and surface courses of flexible pavement were constructed following guideline of Local Government Engineering Department, Bangladesh [11]. The pavements were constructed in adjacent place maintaining the same environmental conditions. Construction procedures have been given in Table 3.

3 Result and Discussion

Dynamic viscosity of BM-3 was greater than BM-1 and BM-2 at 25 °C, but slightly decreased than BM-1 at 90 °C, while shear rates were 25–60 s⁻¹ (Fig. 1a, b). Addition

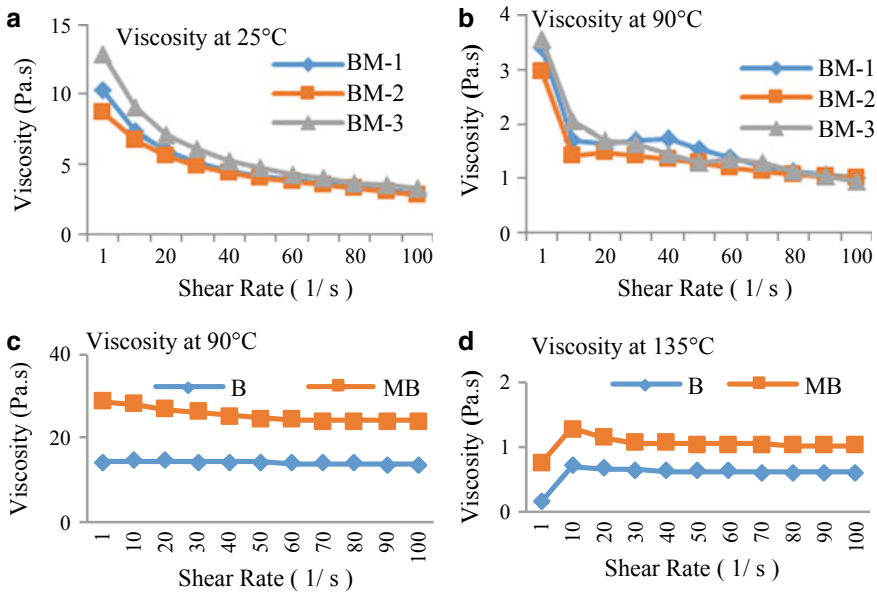


Fig. 1 a Viscosity at 25 °C. b Viscosity at 90 °C. c Viscosity of at 90 °C. d Viscosity at 135 °C

of rubber-additives into BM-1 decreased the mixture viscosity, but increased when soda lignin was added. Dynamic viscosities of modified bitumen (MB) were almost double than 60–70 penetration grade bitumen (B) at 135 °C and 90 °C, as considered for mixing and compaction temperatures, respectively for paving work (Fig. 1c, d). Viscosity decreased with increasing temperature and shear rate in all cases. Tensile strengths and percent elongations of BM-5 and BM-6 were increased than BM-4 (Fig. 2a, b). It was due to curing of natural rubber with additives. The presence of soda-lignin improved the tensile and elongation properties of BM-6 compared to BM-5. So, soda lignin could be used as reactive filler in the curing of natural rubber. While heating of BM-3 with 60–70 penetration grade bitumen (B), the mixture was cured into B and thus specification properties of resulted modified bitumen (MB)

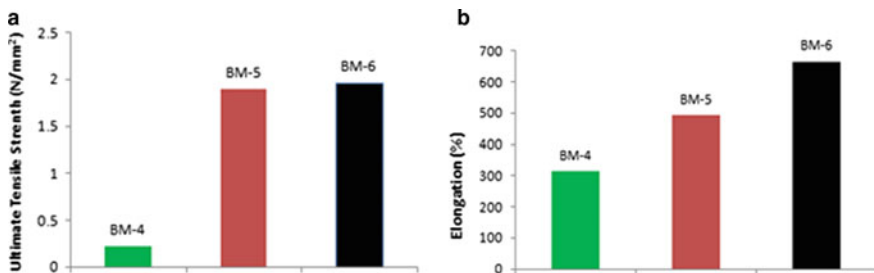


Fig. 2 a Tensile strength. b Percent elongation

Table 4 Specification properties of B and MB sample

Specification properties	B	MB	Method
Softening point (°C)	49	62	ASTM D 36-95
Penetration (1/10th mm)	72	40	ASTM D 05-06
Penetration index	(-) 0.57	(+) 0.87	[10]
Ductility (cm)	96	>100	ASTM D 113-99
Dynamic viscosity@90 °C (Pa s)	14.02	25.20	ASTM D4402
Dynamic viscosity@135 °C (Pa s)	0.61	1.08	
Flash point (°C)	>230	>230	ASTM D 92
Specific gravity@ room temperature	1.02	0.99	ASTM D70-18
Loss on heating (%)	<0.2	<0.2	ASTM D 6

were improved significantly (Table 4). Softening point increased but penetration decreased for the modification. Penetration index increased from (-) 0.57 to (+) 0.87. This may be considered as an indication of low temperature susceptibility of modified bitumen and thus improved resistance to permanent deformation at service temperature. Ductility and other specification properties were also improved due to modification. These properties might be considered as suitable for paving application in tropical countries.

FT-IR data can be considered as a good evidence of internal curing of BM-3 mixture into base bitumen, B. $\text{sp}^2\text{C-H}$ stretching absorption peaks of natural rubber was shown at 3037 cm^{-1} for BM-4 and BM-5, but absent in BM-6. Intensity of this peak was slightly higher in BM-4 than BM-5 due to incomplete curing of natural rubber with additives. BM-6 sample did not show any absorption peak in this area. A broad absorption band at 1661 cm^{-1} confirmed the presence of conjugated C-C double bonds in BM-4 but weak at 1663 cm^{-1} for BM-5. BM-6 showed very weak band at 1653 cm^{-1} . This result can be considered as an evidence of curing of BM-6 in presence of lignin. B showed a broad absorption band at 3279 cm^{-1} due to O-H and/or N-H stretching modes. MB did not show any absorption band greater than 3000 cm^{-1} . After blending of BM-3 with B, resulted MB showed an absorption band at 1719 cm^{-1} . This was probably due to carbonyl stretching of ester group formed during modification, but not observed in case of B. A broad absorption band of sulfoxide was present at 1032 cm^{-1} due to oxidation of B, but very weak at 1033 cm^{-1} as observed in case of MB. Surface micrograph of MB showed homogeneous blend or compatible dispersion with B (Fig. 3a-c). Thermogravimetric analysis showed similar degradation pattern of BM-4, BM-5 and BM-6 samples. No significant weight loss occurred upto $200\text{ }^\circ\text{C}$. Similar thermal stability was also observed for B and MB samples. MB showed slightly better thermal stability compared to B. Marshal stability was improved from 19.1 KN for B to 23.4 KN for MB at 5 (w/w) % additions with aggregates (Fig. 4). It was 25% higher stability than B. No significant stripping happened in case of modified bitumen as observed from static immersion test. Constructed pavements suffered from severe water-clogging during

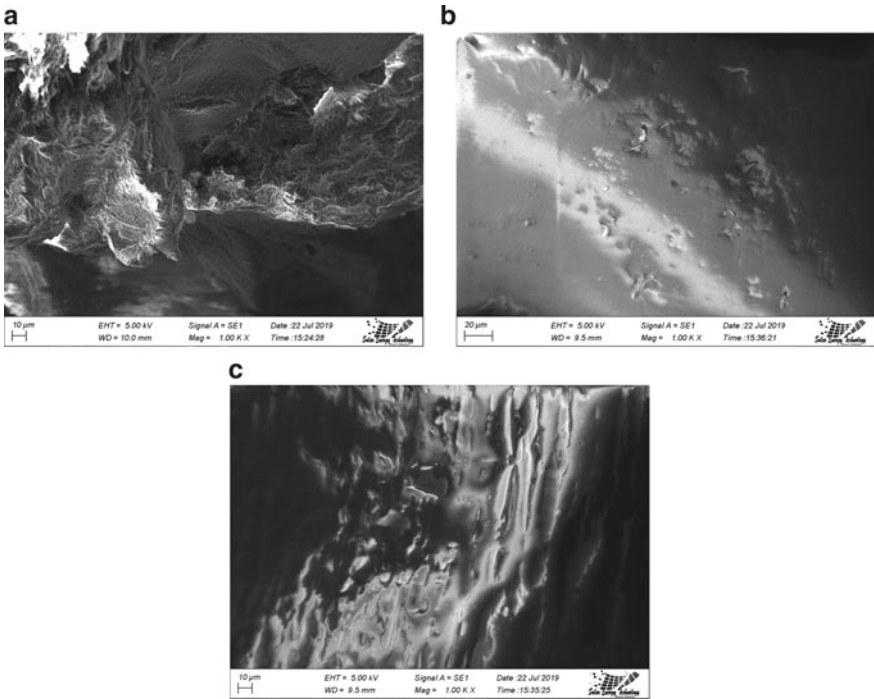
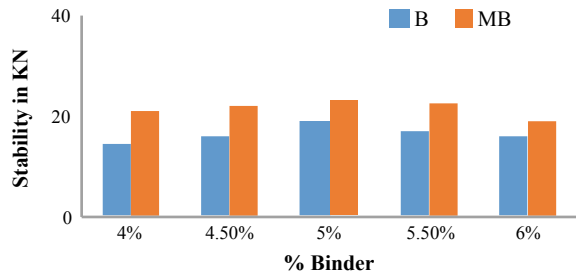


Fig. 3 a SEM of BM-6, b SEM of B, c SEM of MB

Fig. 4 Marshal stability graph



two prolonged rainy seasons (Fig. 5a–c). Pav-MB-PCA showed better performances e.g. water resistance, surface smoothness and durability in comparison to Pav-B-NCA (Fig. 5d, e) as assessed visually. These were due to higher attraction forces between modified bitumen and waste polyethylene coated aggregates.

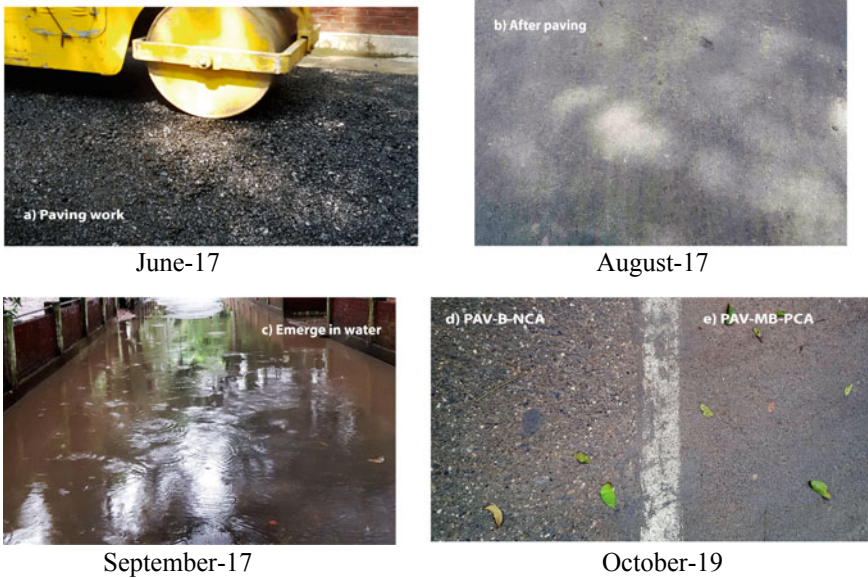


Fig. 5 Pavement construction and surface of the pavements (June-2017 to October-2019)

4 Conclusion

Natural rubber-soda lignin composite could be used to modify 60–70 penetration grade bitumen. Integrated use of modified bitumen and waste thermoplastic coated aggregates for flexible pavement construction was briefly reported in this preliminary work. Field study showed better physical properties e.g. water resistance, heat resistance, surface smoothness and durability of Pav-MB-PCA in comparison to Pav-B-NCA. Detail information on the bitumen modification process and microscopic properties of modified bitumen by atomic force microscopy, scanning electron microscopy, etc. and performance specifications of resulted modified bitumen, recyclability, etc. will be reported shortly.

Acknowledgements This research work was funded by Bangladesh Council of Scientific and Industrial Research, Ministry of Science and Technology, Government of Bangladesh through a development project ‘DFPL-BCSIR (2012–2016)’.

References

1. Willway, T., Baldachin, L., Reeves, S., Harding, M., McHale, M., Nunn, M.: The effects of climate change on highway pavements and how to minimize them. Technical Report 1(124). <https://trl.co.uk/reports/PPR184>. Accessed 20 Nov 2019

2. Pyshyev, S., Gunka, V., Grytsenko, Y., Bratychak, M.: Polymer modified bitumen: review. *Chem. Chem. Technol.* **10**(4s), 631–636 (2016)
3. Zhu, J., Birgisson, B., Kringos, N.: Polymer modification of bitumen: advances and challenges. *Eur. Polym. J.* **54**(1), 18–38 (2014)
4. Vliet, D.V., Slaghek, T., Giezen, C., Haaksman, I.: Lignin as a green alternative for bitumen. In: 6th Eurasphalt & Eurobitume Congress, Prague (2016)
5. Asukar, S.D., Behl, A., Gundaliya, P.J.: Utilization of lignin as an antioxidant in asphalt binder. *Int. J. Innov. Res. Technol.* **2**(12), 198–207 (2016)
6. Israel, A.U., Ogali, R.E., Akaranta, O., Obot, I.B.: Extraction and characterization of coconut (*Cocos nucifera* L.) coir dust. *Songklanakarin J. Sci. Technol.* **71833**(6), 717–724 (2011)
7. Ikeda, Y., Phakkeeree, T., Junkong, P., Yokohama, H., Phinyocheep, P., Kitanodand, R., Kato, A.: Reinforcing biofiller “lignin” for high performance green natural rubber nanocomposites. *RSC Adv.* **7**, 5222–5231 (2017)
8. Košíková, B., Gregorová, A., Osvald, A., Krajčovičová, J.: Role of lignin filler in stabilization of natural rubber-based composites. *J. Appl. Polym. Sci.* **103**(2), 1226–1231 (2007)
9. Sasidharan, M., Torbaghan, M.E., Burrow, M.: Using waste plastics in road construction. Helpdesk Report, K4D (2019). https://assets.publishing.service.gov.uk/media/595_Use_of_Waste_Plast... Accessed 20 Nov 2019
10. Read, J., Whiteoak, D.: *The Shell Bitumen Handbook*, 5th edn. Thomas Telford Ltd, London (2003)
11. Standard Specification for Feeder Road Type-B & Roral Road Type-R1 Under LGED, 1–96 (1999). <https://oldweb.lged.gov.bd>. Accessed 20 Nov 2019