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Feasibility Investigation of Bitumen Properties by Blending of Coal Tar Pitch

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Abstract

There are numerous methods and additives available to improve the durability and quality of road bitumen. A coal tar obtained by coal coking was distilled in a laboratory into fractions of initial boiling point IBP-180˚C (gasoline-like fuel), 180˚C - 360˚C (diesel-like fuel), and >360˚C (residue or coal tar pitch). The coal tar pitch was added into road bitumen by up to 1 - 5 wt% and investigated the alteration of physical and chemical properties. The physico-mechanical properties of coal tar pitch and bitumen blends, as well as the chemical group composition, were determined using standard techniques (MNS) and the SARA method, respectively. Results of 3% coal tar pitch addition into bitumen enhanced ductility by 12.4% and softening point by 1.6˚C. We found that blending with bitumen coal tar pitch as a modifier could improve bitumen properties.

Keywords

Modified Bitumen, Blending, Coal Tar Pitch, Ductility, Softening Point

1. Introduction

Global economic growth causes a rise in vehicle numbers on the road [1]. The traffic problem is worsening, causing major damage to asphalt roadways [2]. The paved road has caused extensive destruction as daily road traffic has increased. As a result, it is critical to extend the ages of asphalt roads, reduce environmental damage, and conduct studies in this field [3]. The properties of the asphalt road are determined by the quality of the asphalt. The various modifiers are utilized for the improvement of the quality of the asphalt. It has several advantages, including durability, adherence to sand and rocks, resistance to deformation, load carrying, and long-term maintenance of attributes in harsh continental climates [1]. Natural and synthetic polymers are employed to enhance the characteristics of asphalt pavements [4]-[9], however, the drawback of this method led to an increment in the cost of bitumen [10].

The main hydrocarbons of bitumen are aromatics (40 wt% to 65 wt%) and resin (30 wt% to 45 wt%). Asphaltenes and saturates contribute 5 wt% - 25 wt% and 0 - 15 wt%, respectively [11]. The physical and rheological properties of bitumen can be determined from the content of hydrocarbon groups [12]. Increasing the content of asphaltenes, which leads to a more stable hardness of the bitumen structure [13]. Bitumen can be classified as sol and gel depending on its hydrocarbon group content. The amount of asphaltenes in the bitumen system reduces as the number of aromatics and resins hydrocarbons increase. As a result, the content of low molecular hydrocarbons in the bitumen system increases the system becomes more unstable. This kind of bitumen is classified as a type of sol bitumen, inversely, bitumen with a high asphaltene content is a gel bitumen [14]. There are some studies to modify the sol type into the gel type, the resin products (phenol-cresol-formaldehyde resin [15] [16] and coal tar [1]) were added in a specified amount to form bitumen with a more stable composition. The different colloidal structures of asphalt are shown in **Figure 1**.

Bitumen is a unique combination of a wide range of chemicals, and it is also challenging to explain the mechanism of bitumen by blending coal tar pitch. Coal tar pitch has been widely explored and employed as a bitumen modifier due to their various functional groups. Low-molecular organic molecules (formaldehyde and maleic anhydride) [17] [18] and sulfur/organic copolymers [19] are generated as byproducts of coal conversion. G. Strap *et al.* [20], and M. Çubuk, *et al.* [21] found out that phenol-formaldehyde resins can be fairly effective petroleum bitumen modifiers. Researcher N. Kamoto *et al.* conducted a study used as an alternative to bitumen by adding 20% of used oil and 50% of crumb rubber to coal tar 30%. The bitumen obtained as a consequence of the study has

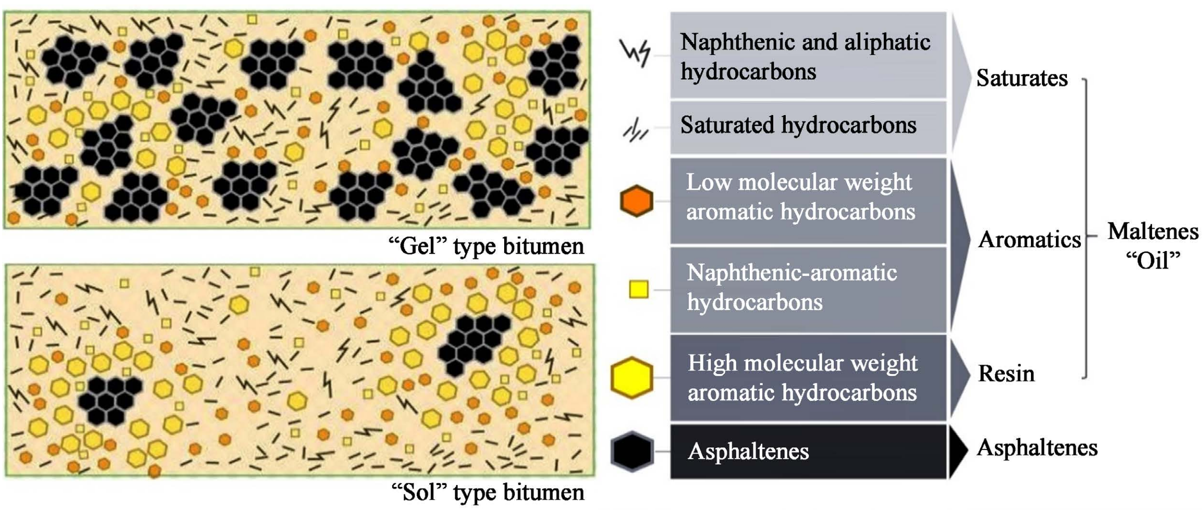

Figure 1. The different colloidal structures of asphalt [12].

low volatility and is resistant to temperature changes [22]. Also, researchers Y. Demchuk *et al.* reported that adding 1% phenol-cresol-formaldehyde resin to bitumen enhances bitumen adhesion [15]. Coal tar pitch has drawn the greatest attention among the various additives due to its excellent modifiers on bitumen quality [23]. Coal tars are byproducts of coal pyrolysis, carbonization, and gasification that are used to produce value-added chemicals and carbon materials [24]. The tar by obtaining coal treatment can be a good binder material [25], and lost its lighter fractions during storage, resulting in asphaltenes [26]. Since coal tar pitch has high contents of resin and asphaltene, it possesses the potential to improve the quality of bitumen [1]. The employment of unmodified bitumen in road asphalt is incredibly sensitive to climate variability [15] and mechanical activities, which leads to the rapid change in the chemical structure of bitumen [10].

Coal tar pitch penetrates into the bitumen system more efficiently than petroleum pitch and enhances adhesion because its functional molecules [27] [28], which include S, N, and O, generate effective physical and chemical interactions with bitumen [29]. Many factors affect the enhancement of bitumen quality by coal tar pitch, including particle size, addition rate, stirring methods, and temperature. Addition of coal tar pitch into the asphalt mixture benefits in the reduction of elastic deformation. During the blending process, light components are converted into a high carbon content as a polycondensation polymer among aromatic rings [1]. Studies on the characteristics of modified bitumen with 5% - 20% coal tar pitch have been conducted [1] [23] and the authors mainly focused on improving the stability and durability of the bitumen system. According to Y. Xue *et al.*, [1], modified bitumen with 15% coal tar pitch enhanced softening temperature by 5˚C while lowering penetration from 88.4 mm to 52.9 mm. This has the advantage of raising the bitumen system's external mechanical force and reducing sun and wind impact, but it also has the drawback of lowering the elasticity of bitumen The main factor affecting the elasticity of bitumen is elongation. During the construction of the bitumen-based road, elongation will determine whether or not cracking will occur. In other words, the greater the stretch, the more it allows the road not to be cracked.

In this study, we investigated the improvement of bitumen properties by blending coal tar pitch obtained by tar distillation, and determined characteristics of modified bitumen by establishing the major parameters such as bitumen penetration, softening point, and elongation. Our study revealed that the addition of 3% coal tar pitch enhanced the ductility and durability of bitumen, which leads to the high probability that the road will not crack. On the other hand, recovering tar residue from coal carbonization as a modifier for bitumen gives the potential to reduce negative environmental effects and waste.

2. Experimental

2.1. Materials

Coal tar obtained by coal coking of Erdenet Mining Corporation in Mongolia was used in this study. This tar was distilled to 360˚C, and the residue or coal tar pitch was employed as a modifier for bitumen. The basic material used in the study was BND90/130 bitumen from Russia.

2.2. Methods of Coal Tar Analysis

The chemical composition of the coal tar was determined by gas chromatograph/mass spectrometer (Agilent 5973N). The tar was distilled into fractions of IBP-180˚C (gasoline-like fuel), 180˚C - 360˚C (diesel-like fuel), and >360˚C (residue or coal tar pitch) [30]. Functional groups of the hydrocarbons of each fraction were investigated by FT-IR (Alpha II, Bruker, Germany).

2.3. Analysis of Pristine Bitumen and Modified Bitumen Characteristics Methods of Coal Tar Analysis

The physical and mechanical properties of pristine bitumen and modified bitumen with various amounts of coal tar pitch were analyzed. MNS 5109-2001, MNS 5211-2002, and MNS 5211-2002 standards were used to assess penetration, softening point, and ductility, respectively. The composition of the hydrocarbon groups of bitumen and modified bitumen was determined by the SARA method [31], which involved precipitating asphaltene with n-heptane in a volume 40 times that of the research sample. Then the maltenes separated from the asphaltene were passed into the Soxhlet apparatus with activated silica gel, and saturated hydrocarbons were extracted by n-heptane, aromatic hydrocarbons by toluene, and resin components by 1:1 toluene-ethanol solution.

3. Result and Discussion

3.1. Analysis of Coal Tar

The chemical compositions of coal tar obtained by coal coking are summarized in **Table 1**.

As shown in **Table 1**, coal tar contains considerable amounts of aromatic hydrocarbons, especially benzene and naphthalene derivatives in a total content of 30.74%. The majority of the chemical composition of coal tar consists of high molecular aromatic and heterocyclic hydrocarbons [32] [33]. However, the composition of coal tar can't be explained completely yet these days. Coal tar includes 20% - 30% phenolic compounds [34] and the main representatives are phenol, (o-, m-, p-) cresol, dimethyl phenol, ethylphenol, methyl ethyl phenol, and trimethyl phenol [35] [36]. The tar analyzed in this study contains about 13.96% of them and nitrogen-containing compounds which are mostly indoles, carbazoles, pyridines, and quinolines [37] and was about 1.66%. The yield of distilled fractions of coal tar is shown in **Table 2**.

The coal tar analyzed in our study has a higher boiling point than 98˚C. Some researchers found out that the IBP of the tar obtained by catalytic hydroprocessing was 118˚C [30] and 159˚C [38]. Furthermore, Maloletnev A *et al.* [39] and Bai Z *et al.* [40] reported that the IBP of the coal tar obtained by distillation was 137˚C and 70˚C, respectively. The IBP of coal tar obtained in different ways is 70˚C - 159˚C in earlier studies, which is near to our results. It can be concluded that tar has less of the compounds which evaporate at low temperatures due to coal tar obtained by the thermal treatment of coal at relatively high temperatures. As shown in **Table 2**, diesel-like fuel represents more than 50% of distilled fractions of coal tar, which is suitable for the processing of basic components in diesel fuel. Some researchers such as Jipeng Meng *et al.* [41], Tao Kan *et al.* [30] and Dong Li *et al.* [42] reported that the yield of diesel fraction is 43% - 64% in coal tar, which is close to our data. All of the fractions obtained from coal tar are valuable and useful, and many researchers have recently focused on the tar residue > 360˚C, which has a yield of 50% - 60% [43].

Figure 2 presents the FT-IR results of distilled fractions obtained by coal tar.

In the IBP-180˚C and 180˚C - 360˚C fractions showed the deformation variation of −OH groups in the 3400 - 3200 (3325 cm^{-1}) absorption ranges, and the alkane ($-CH_2$, $-CH_3$) in the absorption ranges of 2959 cm^{-1} and 2856 cm^{-1}, aromatic C=C valence fluctuations at 1600 cm^{-1}, 1457 cm^{-1}, aromatic C−H deformational fluctuations at 812 cm^{-1}, 752 cm^{-1}, 690 cm^{-1} and, C−O deformational fluctuations at 1243 cm^{-1} 1236 cm^{-1} [44] [45]. As seen in **Figure 2**, three different fractions are complex and with many overlapping bands. The intensity of peaks 3400 - 3200 range (3325 cm^{-1}) corresponding to −OH groups in the above 360˚C fraction significantly decreased compared to other fractions. Here, phenol and its derivatives can be increased in the middle fraction. The main influence of

Table 1. Results of gas chromatograph/mass spectrometer analysis

№	A group of organic compounds	Amount of wt%
1	Aliphatic hydrocarbons	36.08
2	Aromatic hydrocarbons	
	• Derivatives of benzene	10.23
	• Derivatives of naphthalene	20.51
	• Anthracene and Phenanthrene	8.01
3	Phenolic compounds	13.96
4	Nitrogen-containing compounds	1.66
5	Others	9.55

Table 2. The yield of distilled fractions of coal tar.

Results of analysis	
Initial boiling point or IBP (˚C)	98
Fractions of distillation, vol.%	
- IBP-180˚C fraction or gasoline-like fuel)	7.2
- 180˚C - 360˚C fraction or diesel-like fuel)	52.12
- 360˚C above residue or coal tar pitch	40.68

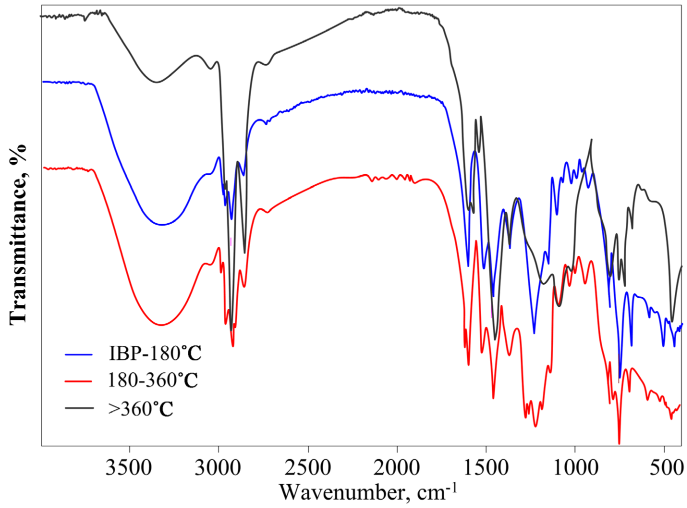

Figure 2. FT-IR spectra for different fractions of coal tar.

light and medium fractions to heavy residues is that the refined absorption of aromatic C=C valence is stronger in the region of 1600 cm^{-1} and 1454 cm^{-1}, which can be explained by the high contents of polycyclic aromatic hydrocarbons in this fraction.

3.2. Physico-Chemical Results of Modified Bitumen

The characteristics of bitumen alter depending on the doped amount of coal tar pitch. Table 3 lists the physical and mechanical parameters of BND 90/130 pristine bitumen and coal tar pitch.

Coal tar pitch obtained by distillation of coal tar was a brittle powdery and black solid material at standard conditions. As shown in Table 3 that the bitumen used in this study has high elasticity, softening point, and penetration, which are within the BND90/130 bitumen standard. Modified bitumen prepared with various amounts of coal tar pitch doped into the bitumen (BND90/130), then determined the basic properties of ductility, penetration, and the softening point. In Figure 3, we can see the influence of coal tar pitch on the ductility and penetration properties of the bitumen.

Our results revealed the addition of 3% coal tar pitch was the highest results than other ones, which could be improved bitumen ductility by 12.4% or 97.9 cm to 110 cm. Bitumen ductility is one of the crucial parameters of bitumen and increases in ductility decrease road break. The addition of tar pitch to bitumen produced favorable effects due to enhancing force of intermolecular adhesion of the imported bitumen. Figure 3 shows the influence of the addition of coal tar pitch on the ductility and penetration of imported bitumen. Results from our study revealed that the appropriate percentage of coal tar pitch was 3%.

As seen in Figure 3, the bitumen penetration reduces as the amount of coal tar pitch increases. The coal tar pitch has a high softening point and solid state at standard conditions, and can greatly improve the hardness abilities of the bitumen system. The softening point evaluates bitumen hardness characteristics, and

Table 3. The basic properties of bitumen and coal tar pitch.

Parameters	Technical requirements	Materials		Standards
		Coal tar pitch	Bitumen (BND90/130)	
Penetration at 25˚C, 0.1 mm	91 - 130	-	91.6	MNS 5109:2001
The softening point, ˚C	>43	76.5	44.7	MNS 5211:2002
Ductility at 25˚C, cm	>65	<0.01	97.9	MNS 5110:2001

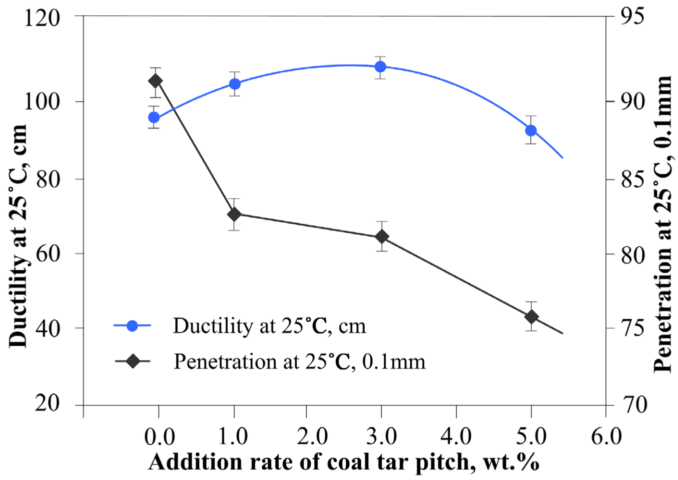

Figure 3. The correlation between penetration and ductility on the addition rate of coal tar pitch.

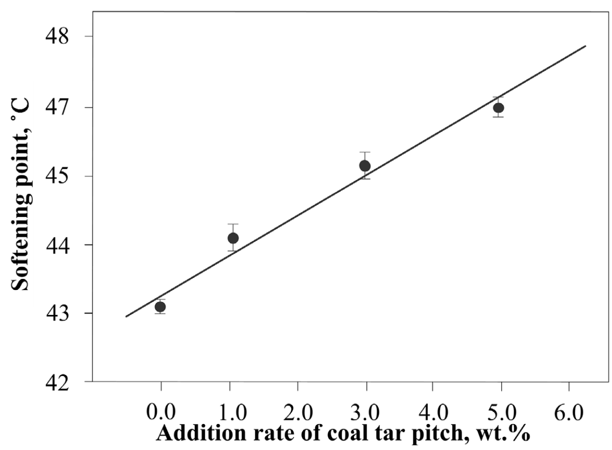

Figure 4. Relation of softening point and the addition rate of coal tar pitch.

Figure 4 shows the relation between the rate of coal tar pitch and softening point.

Figure 4 shows that the percentage of coal tar pitch is directly proportional to the softening point. As the softening point rises, road construction with that bitumen has the advantage of avoiding the formation of the road. The ability to ductility and the softening point of bitumen is directly related to the content of resin, asphaltene, and oil in the bitumen composition. The penetration is a key factor influencing bitumen quality and is disproportional to the value of softening point as can be seen in Figure 5.

Figure 5 shows that by increasing the amount of the coal tar pitch, the penetration decreases while the softening point increases and that these two parameters are inversely related. Physico-mechanical determination is insufficient to evaluate the characteristics of bitumen. Therefore, chemical groups of the bitumen composition and the modified bitumen were determined by the SARA method [31]. The modified bitumen was named modified bitumen 1, 2, and 3 depending on the percentage of the coal tar pitch, respectively, and the results are listed in Table 3.

Table 4 shows that the studied pristine bitumen has low asphaltene contents in its composition. Coal tar pitch contains high concentrations of resin and asphaltene compounds, which can significantly improve bitumen quality. As a result, the addition of coal tar pitch to bitumen enhances adhesion while decreasing weathering [1]. Table 4 shows that the content of resin and asphaltene for the modified bitumen-2 has increased by 1% - 2% compared to the pristine bitumen, however, the content of saturated and aromatic hydrocarbons decreased. Lowering the saturated hydrocarbons in the bitumen system can be decreased its rheological properties or persistent damage caused by environmental and climate impacts, inversely, with increasing asphaltenes in bitumen leads to the improvement of the above mentioned properties and problems [46]. The chemical

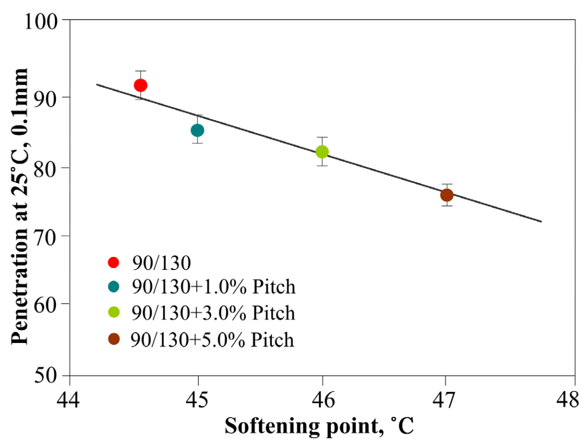

Figure 5. The dependence penetration and softening point.

Table 4. The results of the SARA analysis.

Materials	Addition rate of coal tar pitch, wt%	Saturates, wt%	Aromatics, wt%	Resins, wt%	Asphaltene, wt%
Pristine Bitumen	100	19.1	47.7	24.3	8.9
Modified bitumen-1	1%	18.9	46.9	24.6	9.6
Modified bitumen-2	3%	18.6	45.5	25.2	10.7
Modified bitumen-3	5%	18.1	44.2	26.1	11.6

composition of bitumen is the most important factor in the influence of road deformation. Asphalt-rich bitumen is valuable for that.

4. Conclusion

In this work, we conducted a feasibility study to improve bitumen quality by integrating coal tar pitch created as a coal tar doped into the imported bitumen. According to the findings, we can conclude that adding 3% coal tar pitch into bitumen was the most optimum, with ductility increased by 12.4% as well as softening point increased by 1.6˚C, and coal tar pitch can be an efficient modifier for bitumen.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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