Paving asphalt modifier from co-processing of FCC slurry with coal

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Abstract

Trinidad lake asphalt (TLA) is regarded as a high quality paving asphalt modifier for better road performance and has been used in many countries. However, the high price limits its application to only heavy load surfaces. A co-processing process of fluidized catalytic cracking slurry with coal was developed to produce heavy product (named CS) with asphalt modification ability similar to that of TLA. As a typical example, CS-modified asphalt with CS to base asphalt ratio of 20/80 meets the specifications of ASTM D5710 and BS 3690 for TLA-modified asphalt. The properties of CS-modified asphalt concrete are similar to that of TLA-modified asphalt concrete and satisfy the specifications of JTJ 036-98. Depended on the co-processing conditions used, the yields of the modifier are in the range of 65–80%, with softening point of 90–180 °C and penetration value of 0–8 (1/10 mm) at 25 °C. Asphaltenes content in CS is about 20–70 wt.%, preasphaltenes about 5–30 wt.%, and organic residues bout 5–60 wt.%. Hydrogen content and hydrogen distribution of CS vary with the co-processing conditions. The atomic ratio of H/C of CS is lower than that of TLA.

1. Introduction

Paving asphalt on road experiences a wide range of static and dynamic stress at varying temperatures under different environmental conditions [1]. The limited lifetime of the road surface is due to aging of the paving asphalt. Addition of non-vulcanized nature and synthetic polymers to improve properties of paving asphalt are well known and widely used [2,3]. Reclaimed rubber, pulverized waste tire and special naturally occurring asphalt (such as Trinidad lake asphalt) are also used as additives [4,5]. Addition of these materials improves durability of asphalt surface, adhesion of asphalt to aggregate, deformation resistant at great load and freeze resistance [1–5].

Compared to other additives, Trinidad lake asphalt (TLA) results in super road performance, including exceptional resistance to deformation, low thermal sensitivity and outstanding tenacity under climatic extremes. It is used in pavement applications worldwide, such as in USA, UK, Germany, Japan and China [5,6]. In China TLA was used only in limited places due to its high price ($ 500–750 t^{-1}) and large amounts of addition (25–50%), which result in high capital cost compared to use of other additives [5,6].

Co-processing of coal and low value petroleum fractions has been studied extensively for production of transportation fuels. However, our earlier work showed that the process could be simplified to produce high quality paving asphalts [7–9] as well as high quality paving asphalt modifiers [10,11].

This paper presents detailed experimental data on modification ability and physical and chemical properties of the modifiers produced from the co-processing and compared with that of TLA. The co-processing was made under mild conditions using fluidized catalytic cracking slurry (FCCS) and coal. The effects of the co-processing conditions on the modifier are also discussed. The cost of the co-processing modifier was estimated to be about 30% the market price of TLA.

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2. Experimental

2.1. Materials

A fluidized catalytic cracking slurry, from Shijiazhuang Petroleum Refinery of China, and a Chinese bituminous coal (YZ coal), were co-processed to produce the modifier. The properties of FCCS and the coal are listed in Tables 1 and 2. The base asphalt used to test the modification ability of the modifier is BZ90 (AH 90 in Chinese standard specification for paving asphalt) with a penetration value of 95 (1/10 mm).

2.2. Sample preparation

The co-processing was carried out in a 50-l autoclave with a magnetic stirrer under hydrogen or nitrogen pressures of 8–14 MPa at temperatures of 400–450 °C for 1–3 h. The coal was pulverized to <100 mesh and impregnated with a Fe (1 wt.%) or a Mo (0.04–0.4 wt.%) based catalyst precursor. $FeSO_4$ and urea solutions were used to impregnate Fe catalyst [12], and $(NH_4)_6Mo_7O_{24}$ solution was used to impregnate Mo catalyst. The impregnated coal samples were dried in a vacuum oven under N_2. The ratios of the FCCS to the coal were 1:1 or 2:1 in mass. The 50 liter autoclave was directly connected to a 40-l vacuum distillation unit. After the reaction, the mixture was discharged into the distillation unit and the oil fractions were distilled out at 330 °C and 0.01 MPa. The modifier (named CS) is the residual material collected from the bottom of the distillation unit, which include unreacted and polymerized carbon materials and inorganic materials in the parent coal. Typically the CS yield ranges from 65 to 80% and the oil yield 10 to 25%.

The modification ability of the CS was evaluated by testing the performance of the modified asphalt. The modified asphalt was obtained by mixing the base asphalt with CS or TLA mechanically at a desired temperature. The CS and TLA were pulverized before the mixing. The mixing ratio of the modifier to the base asphalt is 20/80 or 25/75.

2.3. Evaluation of the base and modified asphalts

The standard tests for base and modified asphalt, such as softening point, penetration, and ductility, were carried out following ASTM methods.

The asphalt concrete tests, including Marshal stability and flow (ASTM D1559 and D5581), dynamic stability (rutting test, China standard T0719-1993), limited tensile stain test (low-temperature bending test, ASTM D4123) and freeze–thaw splitting test (AAHTO T283-98), were performed in the Quality Test Center for Paving Asphalt of the Shanxi Research Institute of Communication.

A heavy traffic gradation for asphalt concrete, designated as AC-16I in Chinese Specification, was selected. The gradation and specification limits are given in Table 3. The ratio of asphalt to crushed limestone is 5–100. Marshal specimens were fabricated following ASTM D1559.

Technical Specifications for Construction of Highway Modified Asphalt Pavement (JTJ 036-98), and Specification and Test Method of Bitumen and Bituminous Mixtures for Highway Engineering (JTJ 052-2000), issued by the Ministry of Communications of China, were used.

2.4. Sample characterization

Elemental analysis of the modifiers was carried out on a Vario EL Elemental analyzer. DIGILAB Excalibur FTS 3000MX infrared spectrometer was used for chemical

Table 1
Properties of the FCCS used (wt.%)

Elemental composition	
C	88.97
H	8.12
N	0.48
S	1.35
$H/C_{(atomic)}$	1.10
Group composition	
Saturates	6.7
Aromatics	65.7
Resins	18.7
Asphaltenes	8.9

Table 2
Properties of YZ coal (wt.%)

Proximate analysis	
M_{as}	1.5
A_{as}	4.17
V_{daf}	45.3
Ultimate analysis (daf)	
C	83.88
H	4.77
N	1.17
S	2.67
$H/C_{(atomic)}$	0.68

daf: dry ash free basis; as: as-received; M: moisture; A: ash; V: volatile matters.

Table 3
Gradation used and gradation limits

Sieve size (mm)	Gradation limits	Gradation used	Ratio
19.0	100	100	2.5
16.0	95–100	97.5	15
13.2	75–90	82.5	14.5
9.5	58–78	68	15.5
4.75	42–53	52.5	11.5
2.36	32–50	41	11.5
1.18	22–37	29.5	7.5
0.6	16–28	22	6.0
0.3	11–21	16	5.0
0.15	7–15	11	5.0
0.075	4–8	6	6.0

structure identification. The modifiers were separated into residues (inorganic and organic), preasphaltenes and toluene solubles by solvent extraction. Tetrahydrofuran (THF) and toluene were used as extraction solvents. THF insolubles were defined as residues. THF solubles and toluene insolubles were defined as preasphaltenes.

A thin layer chromatograph coupled with a flame ionized detector (Iatroscan TH-10) was used for group composition analysis of FCCS. Hexane, toluene, and trichloromethane (95%) + methanol (5%) were used to separate the samples into four fractions: saturates, aromatics, resin and asphaltenes.

3. Results and discussion

3.1. Modification ability of CS

Tables 4 and 5 exhibit the modification abilities of CS and TLA to the base asphalt. The properties of the base asphalt, ASTM (American Society for Testing and Materials) and BSI (British Standards Institution) specifications are also given in Table 4 as references. The notation BZ90/CS refers to the modified asphalt by CS and BZ90/TLA to the modified asphalt by TLA. The ratio of modifier to base asphalt is 20/80.

The results in Table 4 show that both CS and TLA have the ability to alter the properties of the base asphalt, increasing the softening point by about 4–8 °C and decreasing the penetration value by 51–54 (1/10 mm) units. In spite of the large decreases in penetration value it is interested to see that the viscous-elasticity of the modified asphalt samples is still good as demonstrated by the high ductility values at 15 and 25 °C, suggesting strong inter-particle and inter-molecular interactions in the mixtures. It is believed that the fine particles from the modifier absorb the high molecular weight components, asphaltenes, in both base asphalt and modifier. This creates new bonds and forms a net-type structure, which in turns increase the viscosity of the mixture [13]. Table 4 shows that the ductility value at 15 °C for BZ90/CS is significantly higher than that for BZ90/TLA, suggesting the superior low-temperature stability of BZ90/CS. This is probably due to the aromatic nature of CS.

The thin-film oven test (TFOT) is used to evaluate the ability of asphalt to resist fast aging under hot mixing conditions. Both the retained penetration (the ratio of penetration values after TFOT to before TFOT) in ASTM D5710 and the decrease in penetration in BS 3690 represent the high-temperature stability of asphalt materials. The high percentage of retained penetration and low decrease in penetration for BZ90/CS sample suggest its superior high-temperature stability.

Table 4 also compares the properties of BZ90/CS and BZ90/TLA samples with ASTM and BSI standard specifications, D5710 and 3690, respectively, designated for TLA-modified asphalt. It clearly indicates that both modified samples meet the specifications.

Table 5 compares the results of asphalt concrete tests, including Marshall stability and flow, high and low-temperature stabilities, and water stability of samples BZ90/CS and BZ90/TLA, which reflect the engineering properties of the asphalt.

The Marshall stability of an asphalt concrete reflects its ability to resist shoving and rutting under traffic. It should be high enough to tolerate traffic adequately, but not higher than the traffic conditions required. The Marshall flow is the ability of asphalt concrete to adjust to gradual settlements and movements in the sub-grade without cracking. The Marshall flow can be considered as an opposite property to the Marshall stability. Both properties should be balanced and meet the required specifications in the same time. The asphalt concrete specimens used in this study were verified for satisfaction on both Marshall properties.

Table 4
Properties of BZ90 and two modified BZ90 (BZ90/CS and BZ90/TLA)

Properties	ASTM D5710		BSI BS 3690		BZ90/CS (20/80)	BZ90/TLA (20/80)	BZ90
	Minimum	Maximum	Minimum	Maximum			
Softening point (ring and ball) (°C)			47	58	48	51.7	44.2
Penetration at 25 °C, 100 g, 5 s (1/10 mm)	40	55	40	60	44	41	95
Viscosity at 135 °C (cSt)	385				–	–	–
Ductility at 25 °C, 5 cm/min (cm)	100				>150	>150	>150
Ductility at 15 °C, 5 cm/min (cm)					101	52	>150
Flash point (°C)	232				>240	>240	>240
Solubility in trichloroethylene (%)	77	90	75	79	88.4	91.2	–
Inorganic (ash) (%)	7.5	19.5	16	19	–	–	–
Retained properties after the thin-film oven test							
Softening point (ring and ball) (°C)					53.6	56.1	48.7
Penetration at 25 °C, 100 g, 5 s (1/10 mm)					36	26	57
Penetration ratio (%)	55				82	63	60
Drop in penetration (%)				20	18.2	36.6	40
Ductility 25 °C, 5 cm/min (cm)	50				144	100	150
Weight loss (%)				0.5	0.26	0.42	0.10

Table 5
Results of the asphalt concrete test

Items	BZ90/CS (20/80)	BZ90/TLA (20/80)
Marshall test		
Stability (kN)	8.9	
Flow (0.1 mm)	22.5	
High-temperature stability (rutting test): dynamic stability at 60 °C, 0.7 MPa (wheel passes/mm)	2000–3500	3100
Low-temperature stability: limited tensile stain ($\mu\varepsilon$) at −10 °C, 50 mm/min	4000	3100
Water stability: freeze–thaw splitting test, tensile strength ratio (TSR, %)	95	99

Rutting test represents the high-temperature stability of the asphalt concrete. Rutting is generated at a wheel path of carriageway in the cases of warmer climate conditions, heavily trafficked roads, approaches to intersections and climbing lanes. Dynamic stability of the asphalt concrete against rutting is an important property of asphalt mixture in wearing course. It represents the ability to resist shoving rutting under traffic and elevated temperature. For modified asphalt concrete this value should be greater than 800–3000 wheel passes/mm depending on the climate encountered [14]. It is clear that the dynamic stabilities of both samples are similar and meet the highest required value.

Limited tensile stain and freeze–thaw splitting tests reflect the low-temperature and water stability. Generally, tensile strength ratio TSR (%), determined by freeze–thaw splitting test should be greater than 75. The limited tensile stain determined by low-temperature bending test should be 2000–2500 under moderate climate conditions, and greater than 3500 under the cold climate conditions. It can be seen that both BZ90/CS and BZ90/TLA concrete meet the specifications. The limited tensile stain for BZ90/CS concrete is significant higher than that for BZ90/TLA concrete, which implies the better low-temperature stability of BZ90/CS. This is consistent with the fact that the ductility of BZ90/CS at 15 °C is significantly higher than that of BZ90/TLA.

3.2. Characteristics of CS and TLA

Table 6 compares the properties of a typical CS and TLA. There is no significant difference in the properties on specific gravity, softening point and penetration value. There

Table 6
Properties of typical CS and TLA samples

Properties	CS	TLA
Specific gravity	1.15	1.39–1.44
Softening point (ring and ball), (°C)	102	93–99
Penetration at 25 °C, 100 g, 5 s (1/10 mm)	8	0–4
Weight loss after thin-film oven test (%)	<1	<2
Solubility in trichloroethylene (%)	53	52–55
Inorganic residue (%)	3	39
Organic residue (%)	20	8
Preasphaltene (%)	33	1
Toluene solubles (%)	44	52
H/C$_{(atomic)}$	0.8	1.4
Aromatic/aliphatic hydrogen ratio by FTIR ($\times 0.01$)	7.4	0

is no significant difference in weight loss after thin-film oven test and solubility in trichloroethylene. There are significant differences, however, in chemical structures and compositions as suggested by the different distribution in inorganic residues, organic residues, preasphaltenes and toluene solubles. Clearly, compared to TLA, CS contains more organic residues and preasphaltenes. CS contains less inorganic residues and toluene solubles. These imply that CS contains more condensed carbon structure. Moreover, if the organic residues and preasphaltenes are considered as dispersed particles (similar to the inorganic residues in TLA), and the group composition of CS and TLA are defined as the toluene solubles and insolubles (where inorganic and organic residues, and preasphaltenes are all toluene insolubles), the group composition distributions of both modifiers are similar. This is consistent with the fact that the modification ability of CS and TLA are similar. Since the organic residues and preasphaltenes have stronger interaction with the base asphalt than the inorganic residue has, CS exhibits better modification ability than the TLA does. The last two lines in Table 6 represent the atomic ratio of H/C determined by elemental analysis and the ratio of aromatic/aliphatic hydrogen by FTIR. It can be seen that most of CS contains aromatic hydrogen structures with less hydrogen content (H/C atomic ratio less than 1); TLA contains almost no aromatic hydrogen structures with more hydrogen content (H/C atomic ratio of TLA is 1.4). These also suggest the more condensed carbon structure of CS.

3.3. Effects of co-processing conditions

Modification ability of CS obtained under different co-processing conditions. Table 7 gives the properties of a series of asphalts modified by different CS samples, obtained under different co-processing conditions. The data of TLA-modified asphalt obtained under the same mixing condition is also listed for comparison. The base asphalt used is the same as above. The ratio of modifier to base asphalt is 25/75. The processing variables include FCCS to coal ratio, catalyst type and loading, reaction temperature, atmosphere, and residence time. It can be seen that the modification ability of CS samples varies with the processing conditions. Comparing with the properties of the base asphalt, the increases in softening point are from 5 to 11%, the decreases in penetration are from 20 to 66% and the decreases in ductility are from >62 to >81%.

Table 7

Modification ability of CS samples produced from different co-processing conditions

Run number	Processing condition (coal:FCCS/catalyst and loading[a]/residence time/ temperature/atmosphere)	Softening point (SP) (°C)	Penetration at 25 °C (1/10 mm)	Ductility at 25 °C (cm)	Flash point (°C)	Retained properties after the thin film oven test				
						SP (°C)	Penetration at 25 °C (1/10 mm)	Penetration ratio (%)	Ductility at 25 °C (cm)	Weight loss (%)
BZ90		44.2	95	>150	>230	48.7	57	60	150	0.100
1	1:2/Fe-1%/1 h/400 °C/N$_2$	47.0	55	38	>260	53.2	33	60	34	0.044
2	1:2/0/1 h/400 °C/H$_2$	46.5	49	40	>260	53.2	32	65	34	0.316
3	1:2/Fe-1%/1 h/400 °C/H$_2$	46.3	67	57						
4	1:2/Fe1/3 h/400 °C/H$_2$	48	51	28	>260	54.3	34	67	41	0.153
5	1:2/Fe-1%/1 h/425 °C/H$_2$	46.2	76	38						
6	1:2/Fe-1%/1 h/450 °C/H$_2$	47.9	59	38						
7	1:1/Fe-1%/1 h/400 °C/N$_2$	47.7	45	34	>260	54.2	32	71	28	0.213
8	1:1/0/1 h/400 °C/H$_2$	47.3	51	40	>260	53.8	38	75	30.2	0.297
9	1:1/Fe-1%/1 h/400 °C/H$_2$	48.9	55	42	>240	56.1	35	64	32.0	0.098
10	1:1/Mo-0.04%/1 h/400 °C/H$_2$	47.7	50	29	>260	55.3	39	72	27	0.250
11	1:1/Mo-0.4%/1 h/400 °C/H$_2$	48.3	32	38	>260	55.1	26	81	29	0.250
12	1:1/Fe-1%/1 h/425 °C/H$_2$	47.1	50	43						
TLA		49.8	46	>150	>250	54.9	31	68	69	0.488

[a] Fe or Mo (wt.%) based on coal feed.

Effects on product distribution of co-processing. Table 8 gives the product distributions of the co-processing under the different conditions. The yields of CS ranges from 65 to 80%, the yields of oil from 7 to 25%. Apparently, the following conditions result in lower oil yields and higher CS yields: (1) without catalyst under H$_2$; and (2) using the Mo based catalyst. The following conditions result in higher oil yields and lower CS yields: (1) with catalyst under N$_2$; (2) extended residence time; (3) higher reaction temperature; and (4) using the Fe-based catalyst.

Effects on softening point of CS. Softening point is an important property for bitumen materials and is crucial for its applicability as an asphalt modifier. The softening points of CS samples obtained under different co-processing conditions range from 90 to 180 °C, which is listed in Table 9. In general, higher temperature, longer residence

time, N$_2$ atmosphere and use of the iron based catalyst produce CS samples with higher softening point. It is believed that under these conditions cracking–condensation reactions are more dominant than that under other conditions.

Effects on hydrogen content and hydrogen distribution of CS. The hydrogen content of the samples is represented by H/C atomic ratio, determined by elemental analysis. The hydrogen distribution is represented by aromatic to aliphatic hydrogen ratio, measured by FTIR. Both (H/C)$_{atomic}$ and (Har/Hal)$_{FTIR}$ ratios for different CS samples are also listed in Table 9. The (H/C)$_{atomic}$ ratios change from 0.65 to 0.85, which are smaller than the ratio for TLA (1.4). The Har/Hal ratios for different CS samples vary from 0.1433 to 0. It is

Table 8

Product distributions under different co-processing conditions (wt.% based on feed)

Run number	Processing condition (coal:FCCS/catalyst and loading[a]/residence time/ temperature/atmosphere)	CS	Oil	Water	Gas + loss
1	1:2/Fe-1%/1 h/400 °C/N$_2$	65.28	24.73	1.00	9.00
2	1:2/0/1 h/400 °C/H$_2$	82.94	7.35	0.71	9.00
3	1:2/Fe-1%/1 h/400 °C/H$_2$	79.84	9.03	2.13	9.00
4	1:2/Fe1/3 h/400 °C/H$_2$	67.41	21.87	1.72	9.00
5	1:2/Fe-1%/1 h/425 °C/H$_2$	71.16	17.16	2.68	9.00
6	1:2/Fe-1%/1 h/450 °C/H$_2$	70.46	17.62	2.92	9.00
7	1:1/Fe-1%/1 h/400 °C/N$_2$	66.56	21.75	2.69	9.00
8	1:1/0/1 h/400 °C/H$_2$	74.50	14.72	1.78	9.00
9	1:1/Fe-1%/1 h/400 °C/H$_2$	76.19	15.29	0.96	7.57
10	1:1/Mo-0.04%/1 h/400 °C/H$_2$	77.63	11.39	1.98	9.00
11	1:1/Mo-0.4%/1 h/400 °C/H$_2$	79.83	9.43	1.74	9.00
12	1:1/Fe-1%/1 h/425 °C/H$_2$	73.24	17.21	2.35	6.81
12b	1:1/Fe-1%/1 h/450 °C/H$_2$	67.28	19.34	3.38	10.00

[a] Fe or Mo (wt.%) based on coal feed.

Table 9

Effects on softening point, hydrogen content and hydrogen distribution of CS samples obtained under different conditions

Run number	Processing condition (coal:FCCS/catalyst and loading[a]/residence time/ temperature/atmosphere)	Softening point (°C)	H/C$_{(atomic)}$	Har/Hal (%)[b]
1	1:2/Fe-1%/1 h/400 °C/N$_2$	124	0.808	10.83
2	1:2/0/1 h/400 °C/H$_2$	101	0.851	6.22
3	1:2/Fe-1%/1 h/400 °C/H$_2$	102.8		6.11
4	1:2/Fe1/3 h/400 °C/H$_2$	90	0.740	9.31
5	1:2/Fe-1%/1 h/425 °C/H$_2$	121.5	0.759	14.33
6	1:2/Fe-1%/1 h/450 °C/H$_2$	127.3	0.833	0
7	1:1/Fe-1%/1 h/400 °C/N$_2$	178	0.769	9.05
8	1:1/0/1 h/400 °C/H$_2$	134	0.822	6.12
9	1:1/Fe-1%/1 h/400 °C/H$_2$	146.3	0.810	6.55
10	1:1/Mo-0.04%/1 h/400 °C/H$_2$	123	0.801	7.28
11	1:1/Mo-0.4%/1 h/400 °C/H$_2$	102	0.827	7.44
12	1:1/Fe-1%/1 h/425 °C/H$_2$	138	0.766	10.70
12b	1:1/Fe-1%/1 h/450 °C/H$_2$	>150	0.649	0

[a] Fe or Mo (wt.%) based on coal feed.
[b] Har/Hal (%) = (aromatic hydrogen peak area/aliphatic hydrogen peak area in IR septum) × 100.

interested to note that the lowest Har/Hal ratio (Har/Hal = 0) is for CS samples obtained at 450 °C and the highest Har/Hal ratio is for CS sample obtained at 420 °C. This may relate to the properties of the coal used. It was found, previously, that the optimum liquefaction temperature is about 400–420 °C for the same coal used in this study [15]. However, the effects of the co-processing conditions on hydrogen content and hydrogen distribution are still not very clear.

4. Conclusions

The heavy products (CS) from co-processing of FCC slurry with coal can be used as paving asphalt modifier. The results from the standard tests for paving asphalt and asphalt concrete show that the modification ability of CS is similar to that of the high price Trinidad lake asphalt. When CS to base asphalt ratio is 20/80, CS-modified asphalt meets the specifications of ASTM D5710 and BS 3690 designated for TLA-modified asphalt. Depending on the co-processing conditions used, the softening point and the penetration value of CS are in the range of 90–180 °C and 0–8 (1/10 mm, at 25 °C), respectively. Asphaltenes content of CS ranges from 20 to 70 wt.%, preasphaltenes from 5 to 30 wt.%, and organic residues from 5 to 60 wt.%. Hydrogen content and hydrogen distribution of CS vary with the co-processing conditions.

Acknowledgments

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