

Restoration of Aged Bitumen Properties Using Maltenes

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Abstract: Reclaimed asphalt pavement (RAP), a material frequently used in asphalt mixtures, is associated with several environmental and economic advantages. Many existing road construction technologies are capitalising on the availability of recycled materials. This study has looked on the usage of maltene as a rejuvenator in aged bitumen. Bitumen with Pen 60-70 was chosen as the control binder. Maltene was added into the aged bitumen at various concentrations (5%, 10%, and 15%) by weight of total binder. The characteristics of rejuvenated bitumen were examined with penetration, softening point, penetration index (PI), viscosity, and storage stability. As the maltene concentration increased, aged bitumen characteristics improved, while the level of stiffness reduced. Mixing temperature and time were significantly linked to the efficiency of blending between aged-virgin binders. Lower mixing temperatures undermined the diffusion of the virgin bitumen into the aged binder, thereby impairing blending efficiency. The results indicate that 15% of maltene can rejuvenate the properties of aged bitumen.

1. Introduction

To achieve the goal of “Zero Waste Management” by 2020, Malaysian institutions of higher education have launched a range of initiatives for managing waste. The majority of which focus on making people more aware of the necessity to cut back on the amount of waste materials disposed in landfills. A key recycling material for the production of a sustainable asphalt mixture is reclaimed asphalt pavement (RAP) [1]. Waste reduction, contribution to the resolution of issues related to discarding of materials from highway construction, counteraction of elevated preliminary expenditure, and conservation of natural resources are the major advantages of RAP [1, 2]. This material is incorporated in the majority of asphalt mixtures currently produced in developed countries. However, unforeseen untimely failure can occur due to the challenges presented by mixtures with high RAP contents to field compact, so RAP can only be added in specific proportions to new mixtures[3]. The restrictions on RAP levels primarily stem from performance problems with the aged binder, which, unlike virgin binder, display greater brittleness and vulnerability to thermal and fatigue cracking [4]. This is due to the natural short and long aging process suffered during service life that lead to increasing the viscosity[5] The chemical composition of the bituminous blend is altered by the existence of RAP binder, yielding a composite asphalt binder of greater hardness and viscosity [6], as the maltenes fraction transforms into asphaltene via oxidation due to the higher viscosity and aging of the RAP binder. However, the



application specification can be met by employing rejuvenators to restore the properties of aged binder. Through supply of more maltenes and/or mediation of improved asphaltene diffusion, rejuvenators ought to ensure that the four chemical constituents of aged binder are in equilibrium [7, 8]. This implies a reduction in viscosity and rigidity and an increase in penetration and ductility [9].

A lot of studies have been conducted to assess the effects of rejuvenators on RAP binders. The research carried out by Zaumanis et al. [10] assessed the efficacy of rejuvenating agents in producing RAP mixtures with 40-100% RAP content. A range of softening agents were used, including waste-derived oils, plant oils, refinery base oils (both non-traditional and traditional), and engineered oils. Additionally, the rejuvenating agents were waste engine oil (WEO), WEO bottoms, WEO with FT wax, distilled tall oil, naphthenic flux oil, organic blend, aromatic extract, refined tallow, and paraffinic base oil. The results demonstrated that each rejuvenating agent resulted in contrasting effects on the aged asphalt binder. The most efficacious in reducing the RAP binder's viscosity was refined tallow. However, WEO bottoms were associated with a higher viscosity level, indicating that they produced greater compaction and mixing temperatures. In the study, the researchers also took performance measurements at low temperatures, the idea being to assess indirect tensile creep compliance and indirect tensile strength. In almost all cases, rejuvenating agents enhanced the mixtures' low-temperature characteristics.

A recent study, conducted by Zhang et al. [11], employed sawdust-derived bio-oil for the rejuvenation of aged asphalt. For the base binders, PG 58-28 and PG 64-22 were used, while the bio-oil concentrations were 10%, 15%, and 20% of the overall binder weight. A range of assessments were performed to gain insight into the aged binders' properties and level of rejuvenation. The results indicated that viscosity and activation energy reduced after adding the bio-oil, whereas temperature susceptibility and viscous component content increased. Additionally, the rejuvenating agent was found to soften the aged asphalt. In terms of the rutting index, this reduced by 75.5% and 77.2% for PAV PG 58-28 and PAV PG 64-22, respectively, at 52-76 °C. Furthermore, the bio-oil improved the low-temperature crack resistance of recycled PAV PG 58-28 and PAV PG 64-22, attaining a standard greater than or equal to that of virgin asphalt. Hence, the rejuvenating agent was effective for PAV PG 58-28 and PAV PG 64-22 at a dose of 15% and 20%, respectively.

Although asphalt binder and rejuvenator recycling has been extensively studied, the rejuvenator potential of maltenes, which is extracted from virgin binder, has not been investigated. Hence, the use of maltenes from virgin asphalt to rejuvenate aged bitumen was examined in the present study. Tests were conducted on penetration, softening point, viscosity, penetration index (PI), penetration-viscosity number (PVN) and storage stability to assess physical properties of rejuvenated bitumen.

2. Materials and Experiments

2.1. Virgin bitumen

The employed control binder was PEN. 60-70 bitumen taken from KBC Malaysia Ltd, which is often used in Malaysian roadworks. Table 1 provides a number of fundamental properties of the bitumen used. Aged bitumen recycling was achieved by using a maltene as a rejuvenator. A chemical solvent and rotary evaporator enabled extraction of the maltene from virgin bitumen.

2.2. Preparation of aged bitumen

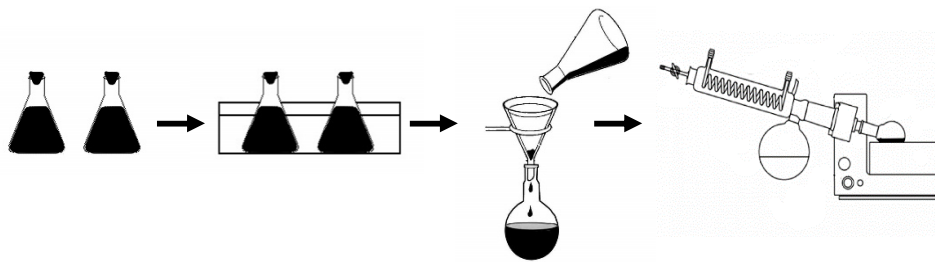
A milling process was performed for sourcing RAP from Secondlink highway with a service life of about 22 years. The bitumen from the RAP was extracted by using methylene chloride solvent as per the ASTM D2172[12]. A rotary evaporator was then used to evaporate out the methylene chloride and recover the aged bitumen as per ASTM D5404[13]. Table 1 shows the properties of aged bitumen. The term applied from this point on is aged bitumen rather than recovered bitumen, for reasons of consistency.

Table 1. Properties of virgin and aged bitumen

	Virgin Bitumen	Aged Bitumen
Penetration (dmm.)	64	7
Softening Point (°C)	50.5	83
Viscosity @ 135 °C (mPa.s)	650	7400
Viscosity @ 165 °C (mPa.s)	200	1500

2.3. Maltene extraction

A multi-stage process was conducted to extract the maltene intended as rejuvenator. The first stage involved mixing virgin bitumen with chemical solvent “petroleum ether” in a flask, which was introduced in a water bath at 50°C and stirred continuously for 120 minutes. In the second stage, the settling of the asphaltenes in the flask was achieved by keeping the dissolved virgin bitumen in a water bath for half an hour, with no stirring. Next, filter paper was employed to attain the fraction of maltene and chemical solvent from asphaltene. Maltene recovery was then achieved with a rotary evaporator at 60-90°C. Finally, complete removal of the chemical solvent was accomplished by placing the maltene in an oven at 110°C for half an hour, as shown in Figure 1. Table 2 provides basic properties of the maltene rejuvenator.

**Figure 1.** Maltene extraction process.**Table 2.** Maltene properties

	Maltene
Density @ 20 °C (kg/cm ³)	0.955
Viscosity @ 60 °C (mPa.s)	782.7
Extracted from:	Virgin bitumen (60-70)

2.4 Samples preparation

The reclaimed bitumen (aged bitumen) was mixed with the virgin bitumen in a proportion of 30% bitumen weight at 145°C and 1500-rpm mixer rotation speed for half an hour. The blending effectiveness of RAP-virgin binders and maltene was substantially influenced by time, temperature and speed. Therefore, the selection of those parameters was based on trial and error. The preliminary study involved blending 0%, 5%, 10% and 15% maltene content by the total binder weight with virgin bitumen that incorporated reclaimed bitumen. The ideal rejuvenator concentration to be added to the mixture was established via penetration, softening point and viscosity tests.

2.5 Experiments

2.5.1. Penetration test. The distance or depth measured in tenths of millimetre permitting achievement of vertical penetration of 100 g weight for a standard needle in a sample at 25°C for 5 seconds is called

penetration. Compliance with ASTM D5/D5M was ensured when the bitumen sample consistency was assessed[14].

2.5.2. Softening point. The thermal sensitivity of the binder is reflected in the softening point, which is assessed in accordance with ASTM D36/D36M[15]. This helps to determine how the binder behaves at raised temperature. The average temperature associated with suitable binder softening and a 25-mm fall of every steel ball prior to coming into contact with the base plate was considered the softening point. The higher the softening point, the less thermally sensitive the binder is.

2.5.3. Viscosity test. In accordance with ASTM D4402/D4402M, the flow resistance and internal friction of bitumen are determined through viscosity measurement[16]. A Brookfield Thermosel viscometer was employed to assess viscosity at 135, 150 and 165°C. Expressed as units of centipoise (mPa.s), low viscosity measurement was reflected in the low friction in the binder sample.

2.5.4. Penetration index (PI). The temperature susceptibility of a bitumen is reflected in the PI [17]. Knowledge of this parameter helps to determine how it will behave in an application. The formula for PI determination is based on equation 1:

$$PI = \frac{(1951.4 - 500 \log P - 20SP)}{(50 \log P - SP - 120.14)} \quad (1)$$

In equation 1, the penetration value (dmm) and softening point value (°C) are respectively denoted by P and SP. A lower PI value indicates a higher temperature susceptibility, with extreme thermal sensitivity being indicated by a PI value of less than -2 [18]. Aged bitumen typically has an elevated PI, suggesting increased brittleness, reduced flow ability and heightened susceptibility to cracking.

2.5.5. Penetration viscosity number (PVN). McLeod N (1976) introduced the PVN as an additional option besides the PI. In accordance with standard specifications for paving asphalt, equation 2 permits PVN determination based on penetration and viscosity at 25°C and 135°C, respectively[19].

$$PVN = (-1.5) \times \frac{(4.258 - 0.7967 \log P - \log V)}{(0.795 - 0.1858 \log P)} \quad (2)$$

In equation 2, the penetration viscosity number, penetration at 25°C and viscosity at 135°C are respectively denoted by PVN, P and V.

2.5.6. Storage stability. In line with ASTM D5892[20], the potential separation of the rejuvenator from the bitumen incorporating RAP under storage was assessed via the storage stability test. Blending was followed straight away by pouring of the rejuvenated bitumen in a 25 mm × 140 mm aluminium tube, which was introduced in the oven for 48-hours heating to 165°C. Sample solidification was afterwards achieved by putting the samples in the freezer at -10°C for four hours. The samples were then separated into three parts of identical length, with the superior and inferior parts being subjected to the softening point test. Control of the discrepancy between these two parts within 2.2°C is recommended.

3. Results and Discussion

3.1. Penetration test

The penetration test was performed on the control sample and bitumen samples rejuvenated with 5, 10 and 15% maltene by weight of total binder. As the maltene concentration increased, penetration increased as shown in Figure 2. More specifically, the penetration increased by 27%, 69%, and 110% as the maltene contents increased by 5%, 10%, and 15%, respectively. One interpretation of these results is a rise in the maltene-to-asphaltene ratio and the softening action of the maltene serving as rejuvenator.

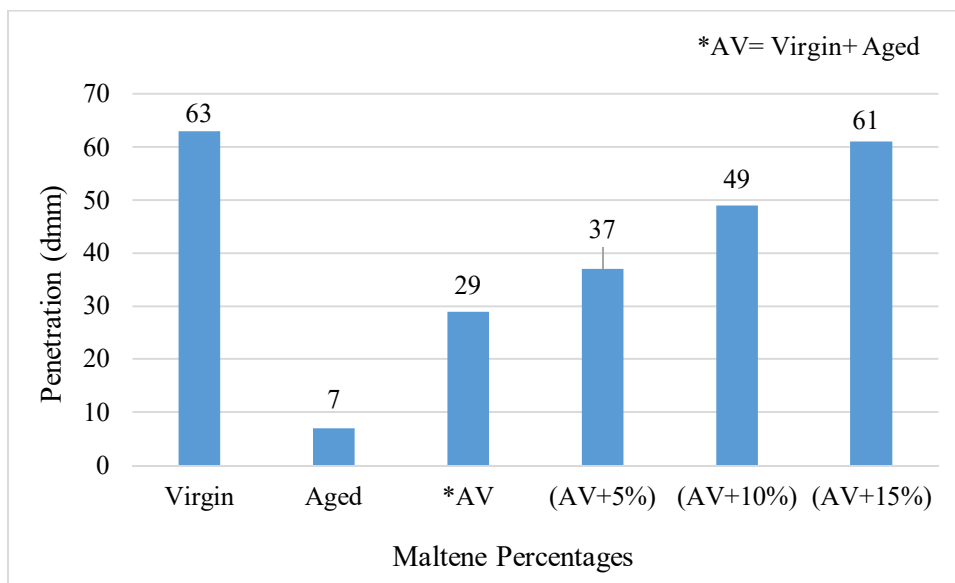


Figure 2. The correlation between penetration and maltene percentages.

3.2. Softening point

Penetration was indirectly correlated with the softening point values of the control binder and bitumen with 5, 10 and 15% maltene rejuvenation (Figure 3). As the maltene concentration increased, the softening point declined markedly. More specifically, a 15% maltene concentration lowered the softening temperature of rejuvenated bitumen to 49°C, which is close to the 50.5°C softening point of virgin bitumen. As a result of such softening point enhancement, pavement degradation owing to fatigue and low-temperature cracking was minimised.

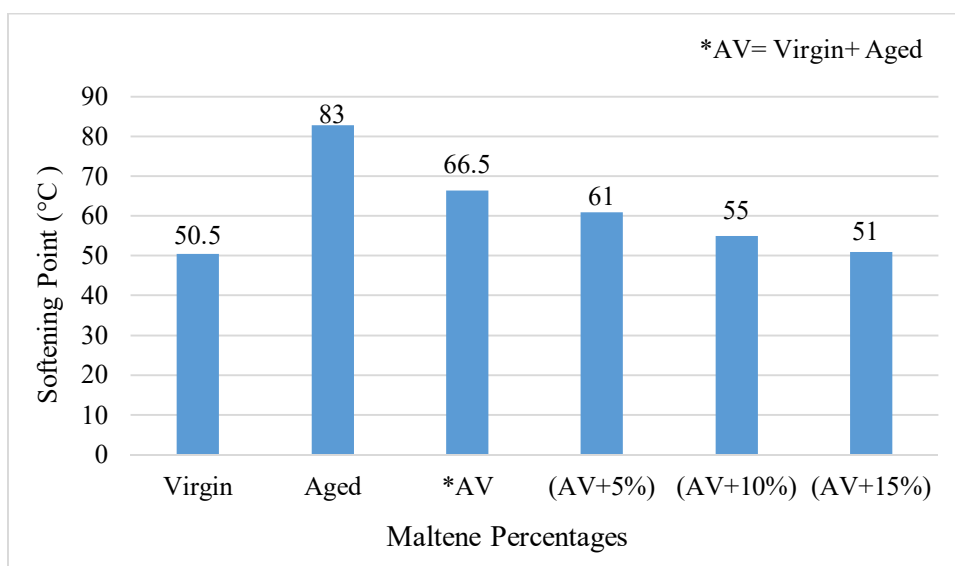


Figure 3. The correlation between softening point and maltene percentages.

3.3. Viscosity test

At 135°C, aged bitumen was more viscous than unaged bitumen, owing to the fact that the binder became markedly more rigid as ageing hardened it and led to high internal friction. Virgin and RAP aggregates must have comparable film thickness and virgin and RAP binder must be homogeneously blended. Therefore, low viscosity is required to achieve sufficient binder flow. The viscosity of virgin, aged and rejuvenated bitumen at various temperatures is indicated in Table 3. Aged bitumen kinematic

viscosity was markedly reduced by 15% maltene, bringing it close to the 650 mPa.s value of virgin bitumen viscosity, as reflected by viscosity at 135, 150 and 165°C. A tight correlation exists between kinematic viscosity and the necessary mixing and compaction temperature.

Table 3. Viscosity of virgin, aged, and rejuvenated bitumen

	Viscosity (mPa.s)		
	135 °C	150 °C	165 °C
Virgin Bitumen	650	400	200
Aged Bitumen	7400	5100	1500
Aged + Virgin = (AV)	1700	800	400
(AV)+5% Maltene	1350	750	400
(AV)+10% Maltene	1050	600	300
(AV)+15% Maltene	675	400	200

3.4. Penetration index (PI)

Thermoplastic properties are possessed by all bitumen types. Whereas high-PI bitumen exhibits softness and is employed in regions with low temperatures, low-PI bitumen displays hardness and is employed in regions with high temperatures. Thus, the behaviour of different bitumen formulations to temperature alterations is quantitatively measured by the PI. Aged bitumen typically has an elevated PI, suggesting increased viscosity and brittleness, reduced flow ability and heightened susceptibility to cracking. This process should be countered by effective recovery of the properties of aged bitumen. Therefore, the quality of rejuvenation may be closely reflected in PI reduction by contrast to extracted bitumen PI. The penetration index of the control, aged bitumen and the bitumen rejuvenated with 5%, 10%, and 15% in this research are presented in Table 4. This proves that the rejuvenated bitumen improved the properties rheological, 15% rejuvenation lowering the aged bitumen PI from 0.96 to -0.99.

Table 4. The correlation between PI and maltene percentage

	PI
Virgin Bitumen	-0.525
Aged Bitumen	0.812
Aged + Virgin = (AV)	0.963
(AV)+5% Maltene	0.508
(AV)+10% Maltene	-0.076
(AV)+15% Maltene	-0.481

3.5. Penetration-viscosity number (PVN)

As it is known, high PVN indicates low thermal susceptibility. Compared to virgin bitumen, aged bitumen had higher PVN, suggesting greater cracking potential. Nevertheless, as shown in Table 5, aged bitumen PVN was markedly reduced by 15% maltene addition, making it similar to virgin bitumen PVN. As a result, cracking-related pavement degradation was minimised. The PVN and PI values were significantly discrepant, without any correlation between them.

Table 5. The correlation between PVN and maltene percentage

	PVN
Virgin Bitumen	-0.04
Aged Bitumen	0.67
Aged + Virgin = (AV)	0.39
(AV)+5% Maltene	0.36
(AV)+10% Maltene	0.34
(AV)+15% Maltene	-0.02

3.6. Storage stability

Separation of rejuvenator and aged bitumen occurs due to the lack of compatibility between binder and rejuvenator. Therefore, rejuvenated bitumen with rejuvenators is morphologically unstable in protracted storage at high temperature. This study investigated the phase separation between maltene and rejuvenated bitumen by conducting the high-temperature storage stability test. This test is indicative of how homogeneously maltene is diffused in bitumen. A bitumen sample is considered to be storage stable at high temperature and a rejuvenator is considered to have good diffusion if the inferior and superior parts have a softening point discrepancy of less than 2.2°C. Table 6 indicates how much the inferior and superior parts of the bitumen samples differed in softening point at 5% (0.5°C), 10% (0.5°C) and 15% (1.0°C) maltene addition. The softening point difference didn't exceed 2.2°C at any maltene concentration. Hence, rejuvenated bitumen constituents retained their stability and homogeneity at high temperature. In addition, the appropriateness of the chosen blending procedure was confirmed by the fact that the maltene-bitumen composite was homogeneously consistent.

Table 6. Storage stability displayed by rejuvenated bitumen

	Softening Point (°C)			
	Top	Bottom	Difference	Requirement < 2.2
(AV)+5% Maltene	63.0	63.5	0.5	Pass
(AV)+10% Maltene	56.5	57.0	0.5	Pass
(AV)+15% Maltene	51.5	52.5	1.0	Pass

4. Conclusion

The results obtained allow the formulation of several concluding remarks. The use of maltene as rejuvenator in 15% concentration enabled the successful recovery the properties of aged bitumen. Penetration was enhanced while the softening point was reduced when maltene being added to aged bitumen as rejuvenator, making aged bitumen less rigid and viscous. Thirdly, the addition of 15% maltene has also decreased PI from 0.96 to -0.48 and PVN from 0.67 to -0.20. This improvement in PI and PVN lead to minimise of cracking-related pavement deterioration. And finally, rejuvenated bitumen constituents retained their stability and homogeneity at high temperature, as confirmed by the high-temperature storage stability test.

5. References

- [1] Soon J L, Serji N A, Nam W P and Kwang W K 2009 Characterization of warm mix asphalt binders containing artificially long-term aged binders *Construction and Building Materials* **23**(6)2371-2379
- [2] Duraid M A, Hussain A K and Riaz A 2018 Novel Methodology to Investigate and Obtain a Complete Blend between RAP and Virgin Materials *Journal of Materials in Civil Engineering* **30**(5) 04018060

- [3] Mohamed E, Christopher W and Eric C 2018 Thermal stability and evolved gas analysis of rejuvenated reclaimed asphalt pavement (RAP) bitumen using thermogravimetric analysis–Fourier transform infrared (TG–FTIR) *Journal of Thermal Analysis and Calorimetry* **131**(2) 865-871
- [4] Alessandro B, Ana J D, Davide L P and Filippo G 2017 Effects of laboratory aging on properties of biorejuvenated asphalt binders *Journal of Materials in Civil Engineering* **29**(10) 04017149
- [5] Cavalli M C, Martins Z, Edoardo M, Manfred N P and Lily D P 2018 Effect of ageing on the mechanical and chemical properties of binder from RAP treated with bio-based rejuvenators *Composites Part B: Engineering* **141** 174-181
- [6] Noferini L, Andrea S, Cesare S and Francesco M 2017 Investigation on performances of asphalt mixtures made with Reclaimed Asphalt Pavement: Effects of interaction between virgin and RAP bitumen *International Journal of Pavement Research and Technology* **10**(4) 322-332
- [7] Elseifi M A, Louay N M and Samuel B C 2011 Laboratory evaluation of asphalt mixtures containing sustainable technologies *Asphalt paving technology* **80** 227-244
- [8] Shen J, Serji A and Jennifer A M 2007 Effects of rejuvenating agents on superpave mixtures containing reclaimed asphalt pavement *Journal of Materials in Civil Engineering* **19**(5) 376-384
- [9] Ali A W, Yusuf A M, Aaron N, Caitlin P and Thomas B 2016 Investigation of the impacts of aging and RAP percentages on effectiveness of asphalt binder rejuvenators *Construction and Building Materials* **110** 211-217
- [10] Martins Z, Rajib B M, and Robert F 2013 Evaluation of rejuvenator's effectiveness with conventional mix testing for 100% reclaimed Asphalt pavement mixtures *Transportation Research Record* **2370**(1) 17-25
- [11] Zhang R, Zhanping Y, Hainian W, Mingxiao Y, Yoke K Y and Chundi Si 2019 The impact of bio-oil as rejuvenator for aged asphalt binder *Construction and Building Materials* **196** 134-143
- [12] ASTM D2127 2017 Standard Test Methods for Quantitative Extraction of Asphalt Binder from Asphalt Mixtures (ASTM International, West Conshohocken, Pa)
- [13] ASTM D5404/D5404M 2012 Standard practice for recovery of asphalt from solution using the rotary evaporator (ASTM International, West Conshohocken, Pa)
- [14] ASTM D5/D5M 2013 Standard test method for penetration of bituminous materials, (ASTM International, West Conshohocken, Pa)
- [15] ASTM D36/D36M 2014 Standard test method for softening point of bitumen (ring-and-ball apparatus), (ASTM International, West Conshohocken, Pa)
- [16] ASTM D4402/4402 M 2015 Standard test method for viscosity determination of asphalt at elevated temperatures using a rotational viscometer (ASTM International, West Conshohocken, Pa)
- [17] Chen J S, M C Liao, and C H Lin 2003 Determination of polymer content in modified bitumen *Materials and structures* **36**(9) 594-598
- [18] Read J, David W and Robert N H 2003 The shell bitumen handbook (*Thomas Telford*)
- [19] McLeod N 1976 Asphalt cements: pen-vis number and its application to moduli of stiffness *Journal of Testing and Evaluation* **4**(4) 275-282
- [20] ASTM D5892-00 2015 Standard Specification for Type IV Polymer-Modified Asphalt Cement for Use in Pavement Construction (ASTM International, West Conshohocken, Pa)

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