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Physical and rheological properties of acrylate–styrene–acrylonitrile modified asphalt cement

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HIGHLIGHTS

- ASA effects on the physical and rheological properties of asphalt cement studied.
- Samples prepared using melt-blending technique at concentrations of 3%, 5% and 7%.
- ASA improved temperature susceptibility of modified asphalt cement.
- Concentration up to 5% of ASA able to increase percentage of the crystalline phase.
- ASA modified asphalt cement resist rutting and fatigue parameters significantly.

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ABSTRACT

Polymer-modified asphalt cement (PMAC) has been used in the last few decades to mitigate several root causes of asphalt-cement failures. Furthermore, it has been proved that polymers used as asphalt-cement modifiers had significant influence on the properties of asphalt at low and high temperatures. The primary objective of this study was to investigate the rheological, mechanical, and physical properties of acrylate–styrene–acrylonitrile (ASA) PMAC. Three types of samples with different concentration of the additives were studied, namely asphalt cement with 3%, 5%, and 7% of ASA. The influence of the modifier on the rheological, mechanical, and physical properties was evaluated by conventional tests (penetration, softening point, and ductility), viscosity, X-ray diffraction (XRD), and measurements from a dynamic shear rheometer (DSR). From the results of this study, it is evident that the addition of ASA has a significant effect on the rheological properties of asphalt cement. The temperature susceptibility of modified asphalt cement (MAC) was reduced compared with unmodified asphalt cement. In addition, the storage-stability test confirmed that the ASA–MAC has good compatibility. Furthermore, XRD results revealed that the phase of ASA–MAC changed from amorphous to semi-crystalline, which indicates that these materials have good workability. Based on the results from the DSR measurements, ASA–MAC has reduced temperature susceptibility, and increased stiffness and elastic behavior in comparison to unmodified asphalt cement. Moreover, ASA–MAC shows improved rutting resistance at high temperatures and higher fatigue performance at low temperatures. As a result, ASA can be considered as a proper alternative additive to modify the properties of asphalt cement.

1. Introduction

Asphalt cement (AC) is mostly used as binder of road surface. The performance of AC significantly depends on its susceptibility upon changes in temperature [1,2]. In various cases, heavy traffic loading and severe weather conditions will result in serious damage of the asphalt-road surface [3,4]. Unfortunately, AC becomes liquid at higher temperatures and is inelastic at lower temperatures, which can result in rutting at high temperatures and

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cracking at low temperatures. This temperature susceptibility limits its application and, therefore, it is essential to enhance the performance of AC by the addition of various modifiers, such as rubber, polymer, and nanoparticles [5–7]. In recent years, polymeric materials are commonly used as additives for modifying asphalt-road surfaces [4,8], thereby mitigating numerous main causes of asphalt-pavement disasters that occurred over the last years [9–12]. These polymers were reported to improve the properties of bitumen, such as its stiffness at high temperatures, cracking resistance at low temperatures, moisture resistance, and fatigue life [13,14]. In homogeneously modified asphalt cement (MAC), the polymer is mingled with AC thoroughly, thereby raising the resistance against fatigue, rutting, as well as aging at medium and high temperatures [15,16]. Moreover, the polymer in MAC should not lead to a highly viscous mixture at elevated temperatures or too a high stiffness at low temperatures. The stability during storage and transportation of polymer-modified asphalt cement (PMAC) must be good in order to obtain better mechanical properties than base AC [17]. The added polymer can strongly enhance the properties of AC, facilitate the construction of safer roads, and the reduction of maintenance costs [18]. The addition of polymers to bitumen is known to impart enhanced service properties, such as improved thermo-mechanical resistance, elasticity, and adhesivity [19]. The polymeric materials used as modifiers of AC can be divided into two categories, elastomers and plastomers. Elastomer modifiers of asphalt extend both low and the high service temperatures, whilst plastomers are known as efficient modifiers at high service temperatures [20–22]. Plastomers increase the viscosity and stiffness of bitumen by forming rigid network structures resisting deformation, while elastomers improve the elastic behavior of bitumen since they resist permanent deformation under tensile forces and recover their original shape after loading [23,24]. The most commonly used elastomers are styrene–butadiene–styrene (SBS) and ethylene–vinyl acetate (EVA) [25–27]. In the present study, the application of acrylate–styrene–acrylonitrile (ASA) tri-copolymer to modify AC is demonstrated and the performance of ASA–MAC in terms of conventional characteristics, temperature susceptibility, storage stability, and rheological properties is investigated.

2. Experimental

2.1. Materials

The base AC was of 60/70 penetration grade, supplied from a factory in Port Klang, Malaysia. ASA white powder, procured from Shijiazhuang Chanchiang Corporation in China, was used as a modifier for AC. The physical properties of the base AC and the modifier ASA are listed in Table 1.

2.2. Sample preparation

Samples were prepared using the melt-blending technique. The ASA modifier was used for the preparation of three differently modified AC samples (excluding the base asphalt cement) at concentrations of 3%, 5%, and 7%. The asphalt was melted and a Silverson high-shear mixer was used for the mixing at a constant temperature of 170 °C (±1 °C) and speed of 5000 rpm for 90 min to produce homogenous mixtures. Moreover, the homogeneity of mixtures evaluated using softening point test. The samples of softening point were taken every 30 min during mixing time 2 h until the value of softening point becomes stabilized then sure the mixtures were homogeneous.

Table 1
Physical properties of base AC and ASA modifier.

Material	Properties	Unit	Limits	Value
Bitumen 60/70	Specific gravity	–	1.00–1.05	1.03
	Penetration @ 25 °C	0.1 mm	60–70	70
	Softening point	°C	47 minimum	47.0
	Viscosity @ 135 °C	(Pa s)	3 maximum	0.5
	Ductility @ 25 °C	cm	100 minimum	⩾100
ASA	Size	mm	–	2
	Specific gravity	–	–	1.04–1.07

2.3. Experimental procedures

2.3.1. Physical properties

Conventional tests, such as penetration (ASTM D5), softening point (ASTM D36), and ductility (ASTM D113) measurements were done according to ASTM specifications to ensure reproducibility and to evaluate the property changes of MAC in comparison to those of the base AC. The penetration value is a measure of the stiffness of AC while the softening point corresponds to the temperature at which AC starts to become a fluid. Ductility is identified as the maximum distance to which a briquette specimen can be elongated without breaking under increasing tensile forces at a specified rate and temperature.

2.3.2. Temperature susceptibility

AC is a thermoplastic material, its consistency changes with temperature. The temperature susceptibility is the change in rheology of AC with changing temperature and is a very important property, as asphalt behavior depends on temperature and rate of loading. Two different ways for determining the temperature susceptibility of AC are being used currently; the penetration index (PI) and the pen-vis number (PVN). PI and PVN are measured according to the following two equations [28,29]:

$$\mathrm{PI} = \frac{1952 - 500\log\ \mathrm{pen} - 20\mathrm{softening\ point}}{50\log\ \mathrm{pen} - \mathrm{softening\ point} - 120} \quad (1)$$

Pen is the penetration value measured at 25 °C.

$$\mathrm{PVN} = \frac{\log L - \log X}{\log L - \log M}(1.5) \quad (2)$$

Here, $\log X$ = the logarithm of the viscosity in centistokes, measured at 135 °C, $\log L$ = the logarithm of the viscosity at 135 °C for the PVN of 0.0, and $\log M$ = the logarithm of the viscosity at 135 °C for the PVN of −1.5. However, the following equations (based on least-squares fits) can be used to calculate more accurate values of L and M.

The equation for the line representing PVN = 0.0 is:

$$\log V = 4.25800 - 0.79670\log P \quad (3)$$

The equation for the line representing PVN = −1.5 is:

$$\log V = 3.46289 - 0.61094\log P \quad (4)$$

Here, V = viscosity in centistokes at 135 °C and P = penetration at 25 °C.

2.3.3. Viscosity

A Brookfield rotational viscometer was used to measure the rotational viscosity of the AC samples, as per ASTM D4402 and AASHTO T316. About 10.5 g (±0.5 g) of base AC were tested to obtain the viscosity by using a spindle 21 with a constant speed of 20 rpm. In this study, various temperatures were applied to inspect the variations of base AC and ASA–MAC. Specifically, the testing temperatures were 100, 110, 120, 135, 145, 155, 165, 175, 185, 195, and 200 °C.

2.3.4. Storage stability test

The storage stability of MAC was measured as follows. The samples were poured into aluminum-foil tubes; the size of the tube being 16 cm in height and 3 cm in diameter. The foil tubes were closed and stored vertically at a temperature of 163 ± 5 °C in an oven for 48 h. Subsequently, the samples were cooled to room temperature and divided into three equal parts. The samples taken from the upper and lower sections were used to assess the storage stability of the ASA–MAC by determining the corresponding softening points. If the difference between the top and the bottom parts was less than 2.5 °C, the samples were considered to have high storage stability. If the softening points differed by more than 2.5 °C, the ASA–MAC was considered to be unstable [30,31].

2.3.5. X-ray diffraction (XRD)

An X-ray diffractometer type Bruker AXS D4 Endeavor was used to investigate the crystal and microstructure of base AC, ASA and ASA–MAC samples with different concentrations of the polymer. Therefore, XRD provides insight into the microstructure of asphalt materials and information about the crystalline principles like layer diameter, interlamellar distance, number of lamellars, height of the unit cell, and aromaticity [32].

2.3.6. Dynamic shear rheometer (DSR)

The dynamic shear rheometer (DSR) is used to characterize the viscous and elastic behavior of AC at high and intermediate service temperatures. Moreover, DSR measures the complex shear modulus G^* and phase angle δ of AC at the desired temperature and frequency of loading, as per AASHTO T315. Frequency sweeps