Applied Clay Science 104 (2015) 1–7

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

Research paper

Storage stability and rheological properties of asphalt modified with waste packaging polyethylene and organic montmorillonite

Ruien Yu [a,b], Changqing Fang [a,b,*], Pei Liu [b], Xiaolong Liu [b], Yan Li [a,**]

[a] School of Mechanical and Precision Instrument Engineering, Xi'an University of Technology, Xi'an 710048, Shaanxi, P. R. China
[b] College of Printing and Packing Engineering, Xi'an University of Technology, Xi'an 710048, Shaanxi, P. R. China

ARTICLE INFO

Article history:
Received 16 June 2014
Received in revised form 20 November 2014
Accepted 24 November 2014
Available online 5 December 2014

Keywords:
Waste packaging polyethylene (WPE)
Organic montmorillonite (OMt)
Asphalt
Storage stability
Rheological properties

ABSTRACT

In this paper, asphalt was modified using waste packaging polyethylene (WPE) and organic montmorillonite (OMt) and the storage stability and rheological properties of the resulting material were investigated. Simultaneously, the structure and morphology were also studied in an attempt to explain the changes in the material properties. The results showed that the addition of OMt improved the storage stability of WPE modified asphalt, but does not compromise its excellent high temperature rheological properties. Fluorescence microscopic images of the material revealed that the OMt slenderized and orientated the WPE micro-fibers promoting improved dispersion of the WPE in the base asphalt. Exfoliated OMt layers in the asphalt played a key role in the WPE microfibers' orientation and promoted its uniform distribution in the asphalt. In addition, scanning electron microscopic images showed the potential low temperature properties advantages of WPE modified asphalt.

1. Introduction

Asphalt is a product of the refinement of crude oil and is a black or dark brown viscous liquid, semi-solid or solid at room temperature (Lesueur, 2009 and McNally, 2011). This material is widely used in roofing, road and pavement applications (Barakat et al., 2005 and Lesueur, 2009). However, the molecular weight of asphalt is low, its molecular weight distribution is wide, and the mechanical properties are sensitive to the temperature. Asphalt is embrittled at low temperature and flows easily at high temperature (Polacco et al., 2006). In addition, its elasticity and aging resistance are poor (Polacco et al., 2008; Zhang et al., 2011 and Jasso et al., 2012). With increased economic activity, road traffic volume has increased (Tennøy, 2010) and vehicle loading is heavier (Liu et al., 2014), both of which are taxing the capabilities of the asphalt's properties in modern road construction. To meet these challenges, it is necessary to improve the properties of asphalt. One of the many methods employed to enhance the asphalt's properties is to blend it with polymers to produce polymer modified asphalt (PMA) (Yildirim, 2007 and McNally, 2011). In the history of the asphalt modification, rubber is the earliest modifying agent, examples of which are natural rubber (NR), styrene butadiene rubber (SBR) and chloroprene rubber (CR) (Morrison et al., 1994; Lesueur, 2009; Zhang and Yu, 2010 and Liu et al., 2013). Compared to the base asphalt, rubber modified asphalt is a great improvement in terms of low temperature properties, elastic properties and toughness. Polyethylene (PE), ethylene-vinyl acetate (EVA) copolymer or other thermoplastic resin modified asphalt offers better temperature sensitivity and high temperature properties (Yeh et al., 2005; García-Morales et al., 2007 and Fang et al., 2009). Thermoplastic elastomer enjoy the properties of rubber and polymer resin, thus they provide advantages in both high and low temperature properties modification. In the modified asphalt industry, thermoplastic elastomer modified asphalt have a large market share. The most representative of these materials is styrene-butadiene-styrene (SBS) block copolymer modified asphalt (Wen et al., 2002 and Modarres, 2013).

Since the quantity of modified asphalt in road construction is huge, the cost of the modifying agent must be taken into consideration (Navarro et al., 2004 and Guo et al., 2009). Because polyolefin products are widely used in daily life, the cost of this polymer is modest and as a recycled material the cost is even lower. Therefore, recycled polyolefins are an excellent choice for use in asphalt modification, from the standpoints of cost and reduction of “white pollution”. In addition, the complex composition of base asphalt makes it thermodynamic incompatible with a variety of modifying agents, especially PE, which negatively impacts the storage stability of the modified asphalt (Hesp and Woodhams, 1991; Lyu et al., 2002; Pérez-Lepe et al., 2006 and Fang et al., 2013). In previous work (Fang et al., 2012) and the relevant reports (You et al., 2011; Zhang et al., 2011 and Jasso et al., 2012), it has been found that the intercalated or the exfoliated composite structure of nano layered silicate additives play a role in the aging resistance of asphalt. In this paper, the storage stability and rheological properties of

---

* Correspondence to: C. Fang, College of Printing and Packing Engineering, Xi'an University of Technology, Xi'an 710048, Shaanxi, P.R. China. Tel.: +86 29 82312038; fax: +86 29 83212512.
** Corresponding author. Tel.: +86 29 82312038; fax: +86 29 83212512.
E-mail addresses: fcqxaut@163.com (C. Fang), jxxy-ly@xaut.edu.cn (Y. Li).

http://dx.doi.org/10.1016/j.clay.2014.11.033

asphalt modified with waste packaging polyethylene (WPE) and organic montmorillonite (OMt) were studied.

2. Experimental

2.1. Materials

2.1.1. Base asphalt

The base asphalt 90A was obtained from China Petroleum & Chemical Corporation Xi'an branch. Its properties are summarized in Table 1.

2.1.2. Waste packaging polyethylene

After washing and drying, the recycled waste milk bags (principal constituent is linear low-density polyethylene (LLDPE) and low-density polyethylene (LDPE)) were extruded into particles which were used for asphalt modification.

2.1.3. Organic montmorillonite

Nanosized OMt was supplied by Zhejiang Fenghong New Material Co., Ltd (China) which was obtained by the cation exchange reaction of calcium montmorillonite (Ca-Mt) with dioctadecyl dimethyl ammonium chloride (D1821). The product was fine powder with creamy white color, and its density was 1.8 g/cm^3.

2.2. Preparation of WPE/OMt modified asphalt

The high speed shearing method was used to prepare the modified asphalt. The process began by heating 500 g of base asphalt using a universal electric resistance furnace in an iron container until it became fluid. Subsequently, after individually adding the modifying agents into the melted base asphalt, the mixture was sheared using a FLUKO FM300 high shear emulsifier (Shanghai, China) for 1.5 h with the speed of 3750 rpm. In the preparation process, the mixture was stirred to promote uniform heating and a thermometer was used to measure the sample's temperature which was maintained at 150 °C. In addition, the mixture sat quiescent for 10 min after being sheared every 0.5 h to ensure full swelling of the additives. In all the prepared samples, the content of WPE was 4 wt% of the base asphalt, 20 g. The sample with no OMt was used as a control group which was labeled as 1# and the content of OMt in WPE/OMt modified asphalt samples numbers 2–5 was 0.3 wt%, 0.6 wt%, 0.9 wt% and 1.2 wt% of the base asphalt, namely 1.5 g, 3 g, 4.5 g and 6 g, respectively.

2.3. Measurements

2.3.1. Storage stability test

In accordance with ASTM standard D5976, a portion of the prepared modified asphalt sample was transferred into a glass tube which was painted with talc powder spacer. The tube was sealed with a cork and stored vertically in an oven at 163 °C for 48 h, then taken out and placed in a freezer at −2 °C for 12 h. After curing, a small hammer was used to gently break the tube into pieces and then the glass fragments and talc powder spacer were cleaned. When the sample softened at room temperature it was cut horizontally into three equal sections. Finally, the storage stability of the sample was determined by measuring the softening point of the top and bottom sections of each sample.

2.3.2. Rheological characterization

The standardized tests used to characterize rheological properties included penetration (25 °C), softening point, ductility (5 °C) and viscosity (135 °C) which were conducted in accordance with ASTM D5, D36, D113 and D4402, respectively. In addition, the dynamic rheological properties of the modified asphalt was determined using a Bohlin Gemini II dynamic shear rheometer (Malvern Instruments, UK) in accordance with ASTM D7175. Temperature sweeps from 40 to 80 °C at 4 °C/min increments were applied at a fixed frequency of 10 rad/s and variable strain.

2.3.3. Fluorescence microscopy

Polymer particle size and its relative distribution in the asphalt were determined by observing the sampled using a Nikon 80i fluorescent microscope (Japan). This was accomplished by placing a drop of the heated modified asphalt on a glass slide and then pressing it into a thin layer with a cover glass. After cooling, the sample was observed in green excitation light.

2.3.4. Scanning electron microscopy (SEM)

The morphology of modified asphalt was obtained using a JSM-6390A scanning electron microscope (JEOL, Japan) and the samples were coated with gold for 100 s in vacuum using a JFC-1600 fine coater (JEOL, Japan).

2.3.5. X-ray diffraction (XRD)

The interlayer spacing of Mt and OMt was carried out using a Shimadzu XRD-7000 diffractometer (Japan) with Cu target Kα radiation ($\lambda = 0.154$ nm), and 2θ angle scans in the range of 2–10°.

3. Results and discussion

3.1. Storage stability of WPE/OMt modified asphalt

The difference in softening points between the top and the bottom sections of the modified asphalt indicate its storage stability. The smaller the difference value, the better storage stability the modified asphalt gets. Softening point of the top and bottom sections of WPE/OMt modified asphalt are shown in Table 2. Compared with the WPE modified asphalt, the storage stability of WPE/OMt modified asphalt with low OMt content improved significantly, while the increase of the OMt content did not have any positive effect on the storage stability. Fig. 1 shows the fluorescence micrographs of the top and bottom sections of WPE modified asphalt and WPE/OMt modified asphalts with 1.5 g and 6 g OMt. From the Table 2 and the fluorescence micrographs of the top sections, it can be seen that the presence of OMt as well as the amount added had no effect on the top section of WPE modified asphalt and the dispersion of the polymer in the base asphalt. However, in the bottom sections, Fig. 1d represents a sample with higher quantity of

Table 1
Physical properties of 90A base asphalt.

Penetration[a]	Softening point[b]	Ductility[c]	Viscosity[d]
86.1 dmm	51.2 °C	111.7 cm	0.45 Pa · s

[a] Depth of a standard needle (100 g) vertically inserts into the asphalt in 25 °C during 5 seconds.
[b] Records the temperature at which the asphalt starts to flow.
[c] Length of stretching both ends of the standard specimen until fracture in the speed of 1 cm/min and under the temperature of 5 °C.
[d] Evaluated the asphalt in 135 °C according to ASTM D4402.

Table 2
Softening point of the top and bottom sections of modified asphalts.

Sample	OMt content	WPE content	Softening point in top	Softening point in bottom	ΔSP[a]
1#	0	20 g	113.9 °C	72.0 °C	41.8
2#	1.5 g		114.0 °C	79.7 °C	34.3
3#	3 g		112.2 °C	77.3 °C	34.9
4#	4.5 g		113.7 °C	70.5 °C	43.2
5#	6 g		114.0 °C	71.5 °C	42.5

[a] ΔSP means the difference in softening points of the top and the bottom sections of modified asphalts.