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Effects of the Loading Levels of Organically Modified Montmorillonite on the Flame-Retardant Properties of Asphalt

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ABSTRACT: The flame-retardant properties of asphalt for some building applications are very important. In this study, we mainly focused on the influence of organically modified montmorillonite (OMMT) on the flame-retardant and other properties of asphalt in a large content range and explored suitable contents of OMMT for modified asphalts. Modified asphalts with different contents of OMMT from 2 to 15 wt % were prepared by melt blending. The X-ray diffraction results revealed that the intercalated structure was formed in the OMMT-modified asphalt. Rubber processing analysis results indicated the formation of a filler–network structure in the OMMT-modified asphalt. The limiting oxygen index and cone calorimetry results suggested that OMMT could be used as efficient and ideal flame retardants of asphalt. The results also reveal that excess OMMT contents (i.e., >10 wt %) depredated the flame-retardant performance of the modified asphalt. We analyzed the mechanism by taking into account of the features of the modified agent for OMMT. The experimental data showed that the viscosity, softening point, and penetration index of the OMMT-modified asphalt increased with increasing OMMT content, but the ductility decreased slightly when the OMMT content was not beyond 7 wt %. © 2014 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 10.1002/app.40972.

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INTRODUCTION

Asphalt has been widely used in construction areas, such as road tunnels, underground garages, gas station floors, buildings, and waterproof material. Because of its good properties, including low noise, good slip resistance, easy maintenance, and ride comfort, asphalt has become the main material of road construction.[1] In the United States, over 550 million tons of hot-mix asphalt is produced annually for construction projects.[2] However, asphalt is highly flammable and gives out a lot of smoke while burning. What is more, asphalt flows easily only after it is heated to a not very high temperature, so it makes fire spread in a short time. Furthermore, gases (e.g., alkanes, benzene, and methylbenzene) produced during asphalt combustion can not only accelerate combustion, but it can also cause people to choke to death. These hazards limit the application of asphalt in tunnel road projects and make it necessary to change the combustion properties of the asphalt. Some efforts have been made in flame-retardant modifying asphalts; these include the addition of organic flame retardants (e.g., halogen and phosphorus flame retardants) and some inorganic flame retardants (e.g., magnesium hydroxide, aluminum hydroxide). However, environmental or cost problems or the large addition of flame retardants limit some uses of the methods used to modify asphalt.

Because of their large aspect ratio, intriguing multiscale structure, and low cost, montmorillonite (MMT) and other layered silicates, including rectorite, vermiculite, and kaolinite clays, have recently been widely used for the modification of asphalt. Organically modified montmorillonite (OMMT) has a larger layer spacing and forms an intercalated structure or exfoliated structure much more easily to show the better modification effects, so it has many more applications than MMT in asphalt modification in recent years.[3] According to the references, a number of properties of MMT or OMMT-modified asphalt, such as the aging-resistance properties,[4–9] mechanical properties,[10] rheological properties,[11] thermal properties, physical

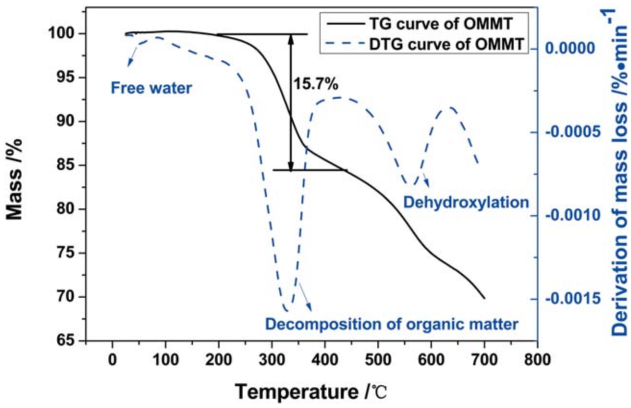

Figure 1. TG and DTG curves of OMMT. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

properties,[4] gas-barrier properties, and water-resistance properties,[12,13] are superior to those of pristine asphalt.[14–16]

Recently, OMMT has been added to polymers for flame-retarding purposes.[17,18] Before the nanocomposite structure collapses, it can help to hinder the permeability of oxygen and heat in the bulk material. After the nanocomposite structure collapses at higher temperatures, MMT is free to migrate to the surface of the material and form a thermal-insulating and low-permeability superficial clay-rich char; this significantly reduces the peak heat release rate (pkHRR) for almost all types of nanocomposites, regardless of the nature of the polymer matrix and the initial nanostructure (intercalated or exfoliated).[19,20]

Considering a similar effect with asphalt, special attention should, therefore, be paid to OMMT-modified asphalt. OMMT, which is a low-cost and environmentally friendly material, has been used to modify asphalt for many purposes. Some researchers have reported the primary results of limiting oxygen index (LOI) of OMMT-filled asphalt, suggesting the potential application of OMMT as a flame retardant of asphalt. Wu et al.[9] reported that the incorporation of 7 wt % OMMT increased the LOI of AH-90 standard asphalt from 19.8 to 20.4. On the other hand, Zhang et al.[21] reported that OMMT showed little effect on the LOI when the OMMT content was below 20 wt %; however, the LOI of bitumen increased from 20.8 to 21.8 when the OMMT content was increased up to 40 wt %. Furthermore, to the authors' knowledge, a comprehensive study on the effect of OMMT on the flame course of the asphalt and smoke release during combustion has not been reported. In terms of the costs and modified effects, the optimization of the OMMT loading level for modified asphalt is also important and necessary. In this study, a large content range of OMMT (2–15 wt %)-modified asphalts was prepared, and we performed a comprehensive performance evaluation, especially with regard to the flame-retardant properties. The structure of OMMT-modified asphalt was analyzed by X-ray diffraction (XRD) analysis and rubber processing analysis (RPA), in which a dynamic mechanical rheometer was used to analyze the filler–network effect of the rubber composites. Through LOI and cone calorimetry (CONE) testing of the asphalt filled with different contents of OMMT, the effect of the OMMT loading level on the flame-retardant asphalt was evaluated. In the meantime, the physical properties of the OMMT-modified asphalt were characterized. According to these experiment results, the mechanism for the effect of OMMT on the flame-retardant and physical properties of the modified asphalt is also discussed.

EXPERIMENTAL

Materials

The asphalt selected for this study, AH-70, was obtained from the Qinhuangdao Petroleum Petrochemical Co., Ltd. The flash point of this type asphalt was 250°C, the ignition point was 330°C, and the LOI was 24.1%. Now, in China, asphalt (AH-70) has been widely used for road pavement in view of its good properties, so in this study, we selected it for our base material and examined its flame-retardant properties when modified by OMMT.

OMMT (Nanomer I.30 P) was produced by Nanocor (Hoffman Estates, IL). The modifying agent of this type of OMMT was octadecyl ammonium salt, and the content was 15.7 wt %; this could be estimated from the thermogravimetric analysis (TGA) results, as shown in Figure 1. TGA was performed on a Mettler-Toledo TGA STARe system under an atmospheric air stream of 10 mL/min, and the TGA experiment was started from room temperature to 700°C at a linear temperature increase of 10°C/min.

Preparation of the OMMT-Modified Asphalt and Testing Samples

The asphalt was first loaded into different stainless steel containers to prevent aging caused by repeated heating of the same sample, which may affect the flame-retardant properties of the asphalt. Each stainless steel container was heated in an oven at 110°C for about 20 min to melt the asphalt inside. The container was transferred to an electric heating oven and maintained at 160°C. After the asphalt was heated up to 160 ± 5°C, a predetermined amount of OMMT (i.e., 2, 5, 7, 10, 12, or 15 wt %) was added to the hot asphalt. The asphalt was stirred with an FM200 shear machine (FLUKO Equipment Co., Ltd., Germany) at 170 ± 5°C for 30 min at a high speed of 5000 rpm for 30 min and then at 2000 rpm for 60 min to expel air bubbles in the asphalt and ensure the homogeneous dispensation of the OMMT.

The 85 × 6 × 4 mm^3 strips test strips for the LOI test and the 100 × 100 × 3 mm^3 test slices for the CONE test were prepared by mold casting according to ref. 1.

Structural Characterization

XRD analysis was carried out on a Rigaku D/max-UltimaIII X-ray diffractometer with Cu Kα radiation (40 kV and 40 mA, $\lambda = 0.154$ nm) at room temperature.

The strain amplitude dependence of the dynamic properties was examined with an RPA2000 rubber processing analyzer manufactured by Alpha Co. By analyzing the relationship between the dynamic modulus (G') and strain, we indirectly obtained the filler–network effect in the OMMT-modified asphalt. The test temperature was 40°C, and the strain scale was 0–400%.

Flame-Retardancy Tests

The flame-retardant performance was tested by a JF-606 oxygen index instrument (Jiangning Analysis Instrument Factory,

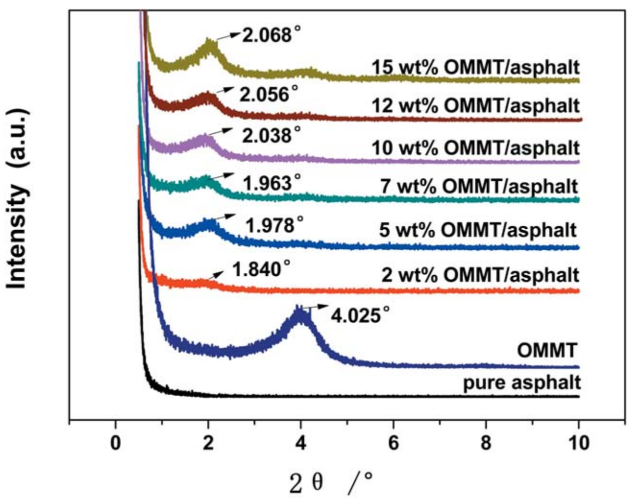

Figure 2. XRD curves of the OMMT and OMMT-modified asphalts with different contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

China) and an FTTO242 cone calorimeter (FTT Co., United Kingdom).

The LOI was measured according to Chinese standard GB/T 2406-2008. During the combustion process, the asphalt test strip showed softening deformation, lodging and dropping phenomenon, and sometimes also a jumping fire phenomenon. These phenomena influenced the accuracy of the results. For better accuracy, each test was repeated at least 12–16 times, and strictly vertical burning of the test strip had to be guaranteed during the combustion. The LOI was calculated with the following equation:

$$\text{LOI}=[O_2]/([O_2]+[N_2])\times 100\% \quad (1)$$

where $[O_2]$ and $[N_2]$ are the volumetric flow rates (L/min) of oxygen and nitrogen gas, respectively. The higher the LOI was, the higher the oxygen volume concentration required to burn was.

CONE analyzes flame retardants through the measurement of oxygen consumption during the fire test, in which the irradiation heat flow is well controlled and maintained. Specimens were wrapped in aluminum foil, which was used to wrap the bottom and sides of the specimen to prevent heat transfer along all of the boundaries except the top burning surface, and then placed in the combustion box. The external heat flux was set 50 kW/m^2.

Physical Property Tests

In accordance with the specification of "Standard Test Methods of Asphalt and Asphalt Mixture for Highway Engineering" (JTJ 052–2000), the viscosity (T0625–2000), the softening point (T0606–2000), the ductility (T0605–1993), and the penetration (T0604–2000) of the OMMT-modified asphalt were measured.

The viscosity of the OMMT-modified asphalt was measured by a DV-II+Pro rotational viscometer (Brookfield, CT). The experimental temperature was 135°C. The rotor model was 21S, and the rotor speed was 20 rpm. The weight of the sample was in the range 8.0–10.0 g. The softening point of the OMMT-modified asphalt was measured with a 20–2200 digital softening point tester (INFRATEST, Germany). The ductility (5°C) of the OMMT-modified asphalt was measured with an H-1068 digital ductility testing machine (Humboldt). The penetration (at 25°C) of the OMMT-modified asphalt was measured with an H-1240 automatic asphalt penetrometer (Humboldt).

RESULTS AND DISCUSSION

Microstructure of the OMMT-Modified Asphalt

XRD Analysis. The XRD curves of the OMMT and different contents of the OMMT-modified asphalt are shown in Figure 2. The diffraction peak of OMMT appeared at $2\theta = 4.025°$. The corresponding interlayer spacing (d) could be calculated according to the Bragg equation ($d = \lambda/2 \sin \theta$), and the result was 2.19 nm. The diffraction peaks of the OMMT-modified asphalt appeared around 2°, and the corresponding d was about 4.41 nm. Among them, the diffraction angle for the 2 wt % OMMT-modified asphalt sample was the smallest. Its diffraction peak appeared at 1.84°, and its corresponding d was about 4.80 nm. The previous results demonstrate that the d of OMMT increased significantly after melt blending with asphalt; this suggested that asphalt molecules could be inserted into the interlayer of the OMMT with formation of the intercalated structure. With increasing OMMT content, the XRD peak shifted to a higher angle; this indicated a decrease in the interlayer height of the intercalated OMMT structure.

RPA Results. Figure 3(a) shows the RPA results of the OMMT-modified asphalt. For a better comparison, the curves of the low-content OMMT-modified asphalt [Figure 3(b)], not the 15 wt % OMMT-modified asphalt sample, was also plotted. The Payne effect in the OMMT-modified asphalt is shown clearly in Figure 3(b). There was a plateau region in the curve at small strain, in which the G' of the OMMT-modified composites was almost unchanged with increasing strain; this suggested that the filler–network structure could not be destroyed within this strain scale. The wider the initial modulus plateau was, the more stable the filler–network was, and the better the filler dispensation was. When the strain exceeded the critical value, the G' of the composites quickly decreased to another plateau; this indicated that the filler–network structure was destroyed.[22] The initial modulus of the composites increased with increasing OMMT content because of the higher degree of the network. As shown clearly in Figure 3(a), the 15 wt % OMMT-modified asphalt showed a quite higher initial G' than the other OMMT-modified asphalt. This suggested a quite high degree of the network and a strong interaction among the fillers. On the other hand, the width of the initial modulus plateau with strain for OMMT-modified asphalt decreased when the OMMT content increased. This implied that the stability of the filler–network and the filler dispersion uniformity of OMMT worsened with the increasing OMMT content. Therefore, 15 wt % OMMT was too much to achieve a good dispersion of OMMT in the asphalt.

Effect of OMMT on the Flame-Retardant Properties of Asphalt

LOI. The results of the LOI test are shown in Figure 4. It can be seen that when the OMMT content was no larger than 7 wt %,

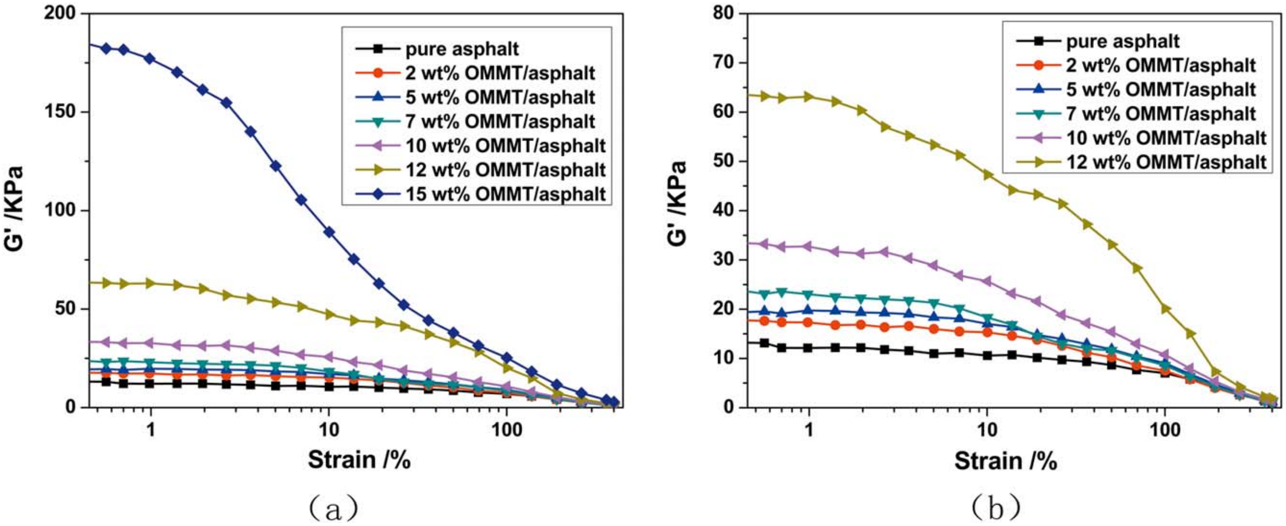

Figure 3. RPA curves of the OMMT-modified asphalts. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the LOI value of the OMMT-modified asphalt increased from 24.1 to 31.1% with increasing OMMT content. Then, the LOI value remained at about 31% when the OMMT content was between 7 and 10 wt %. The LOI dropped to 28.5% when the OMMT content was 12 wt % and continued to drop to 22.3% when the OMMT content was 15 wt %. These results demonstrate that the incorporation of an appropriate amount of OMMT could dramatically improve the flame-retardant performance of the asphalt. During the test, with increasing OMMT content, the phenomenon of softening deformation, lodging, and dropping of the OMMT-modified asphalt weakened.

When the LOI value of a material is higher than 27%, the material is a self-extinguishing material. According to Japanese Standard JISK 7201 requirements, flame-retardant materials are divided into five categories, as shown in Table I.[23] In this study, the LOI of pure asphalt was 24.1%, so the pure asphalt could be classified as nonflame-3. When the OMMT content was 7 wt %, the corresponding LOI was 31.1%, and this material could be classified as nonflame-1, that is, a material having the highest flame-retardant capability. So, we could draw a conclusion that OMMT should be an efficient and ideal agent for increasing the LOI of asphalt, compared with other types of environmentally friendly flame retardants, such as magnesium hydroxide[1,24] and aluminum hydroxide.[25]

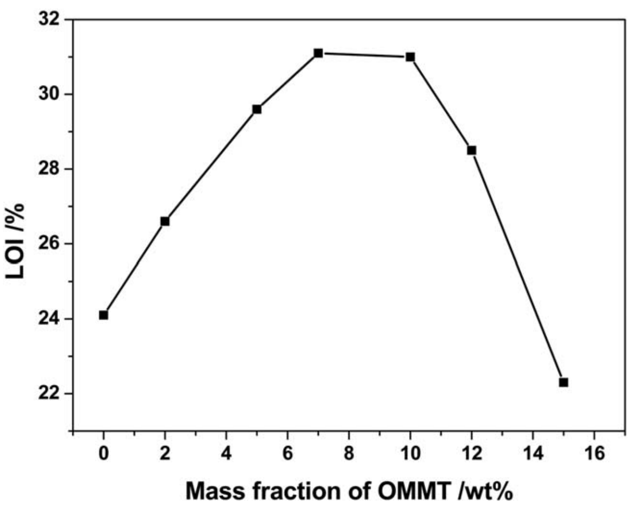

Figure 4. Plot of the LOI of the asphalts as a function of the OMMT content.

CONE. Four modified asphalt samples with different OMMT contents (i.e., 0, 2, 5, and 10 wt %) were chosen to conduct CONE testing. A number of important fire properties, including the time to ignition (TTI), heat release rate (HRR), pkHRR, total heat release (THR), and total smoke release (TSR), were evaluated.

The HRR curves, which describe the time trend of the HRR of the burning asphalt during the whole test, are shown in Figure 5. The prominent combustion peak for pure asphalt disappeared, and the HRR remained relatively constant during combustion after the addition of OMMT. The addition of 2 wt % OMMT obviously reduced the value of HRR throughout the combustion process. The curve of 5 wt % OMMT-modified asphalt showed that the combustion peak value was lower than that of the 2 wt % OMMT-modified asphalt. However, when the burning time exceeded 250 s, the HRR value of the 5 wt % OMMT-modified asphalt was higher than that of 2 wt % sample; and after 300 s, the HRR value of the 5 wt % OMMT-modified asphalt was even higher than that of pure asphalt. The total burning time of the 5 wt % OMMT-modified asphalt was slightly extended compared with that of the pure asphalt. The combustion peak value of the 10 wt % OMMT-modified asphalt was a bit higher than that of the 5 wt % sample, but it was still lower than that of the 2 wt % one. However, the total burning time of 10 wt % OMMT-modified asphalt was greatly extended by about 80% compared with other samples.

In the CONE test, TTI referred to the time from the beginning of surface heating to the continuous burning of the sample at a

Table I. Regulations of Japanese Standard JISK 7201

Level	Nonflame-5	Nonflame-4	Nonflame-3	Nonflame-2	Nonflame-1
LOI (%)	LOI < 21	21 < LOI < 24	24 < LOI < 27	27 < LOI < 30	LOI > 30

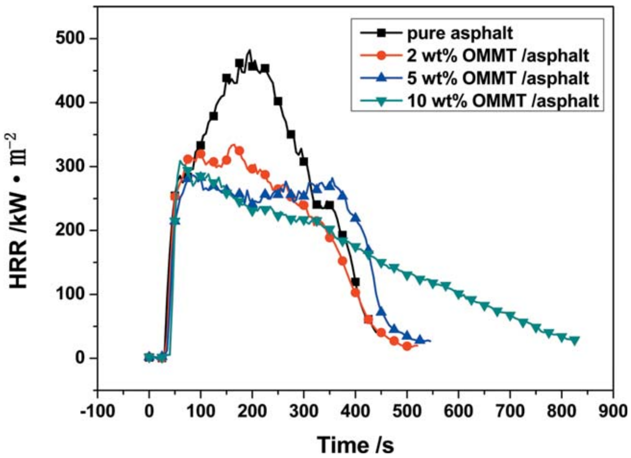

Figure 5. HRR curves of the CONE tests for the pure asphalt and OMMT-modified asphalts with different contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

given thermal irradiation. The longer the TTI was, the better the flame-retardant properties of the material were.

The HRR is another important indicator of flame-retardant performance. The larger the values of HRR and pkHRR were, the larger the heat release of the burning course of the material and the greater the fire hazard were. The pkHRR value was strictly related to the thermal properties of the specimen during the burning process, such as the thermal conductivity and heat capacity.[19] $MHRR_{360}$ is the mean heat release rate burning within 360 s. This time interval of 360 s has a close relationship to the fire safety evacuation time, so $MHRR_{360}$ is of great significance for fire rescue. The lower the pkHRR and $MHRR_{360}$ values were, the better the flame-retardant effect of the OMMT-modified asphalt was. According to the curves displayed in Figure 5, the TTI, pkHRR, and $MHRR_{360}$ values of the OMMT-modified asphalt with different contents could be estimated, as summarized in Table II. We found that the TTI generally increased with increasing OMMT content. The incorporation of 2 wt % OMMT resulted in dramatic decreases in the pkHRR and $MHRR_{360}$ values. With further increases in the OMMT content up to 10 wt %, the modified asphalt exhibited lower values of pkHRR and $MHRR_{360}$, but the changing degree with OMMT content was obviously slight. Moreover, the pkHRR value of 10 wt % OMMT-modified asphalt was a little higher than that of the 5 wt % sample. After all, the higher the OMMT content was to some extent, the better the fire safety performance of the OMMT-modified asphalt was.

Table II. TTI, pkHRR, and $MHRR_{360}$ Results for the OMMT-Modified Asphalts

Sample	TTI (s)	pkHRR (kW/m^2)	$MHRR_{360}$ (kW/m^2)
Pure asphalt	26	482.1	333.4
2 wt % OMMT/asphalt	30	334.6	256.6
5 wt % OMMT/asphalt	33	286.3	251.2
10 wt % OMMT/asphalt	37	308.9	232.9

The THR is defined as the integral of the HRR curve with respect to time; thus, it represents the heat evolved up to an assigned point. The value of THR is almost not influenced by other external factors under the condition of sufficient ventilation. Therefore, THR is considered as an important index for evaluating the fire safety of materials. The higher the THR value of the burning material is, the higher the fire hazard of it is. Figure 6 shows that the addition of OMMT reduced the value of THR. With increasing OMMT content, the value of THR decreased in early stage of the combustion process. When the burning time was prolonged, the THR values of the 5 and 10 wt % OMMT-modified asphalts become larger than that for the 2 wt % sample. The THR value of the 10 wt % sample at the end of burning course was almost same as that for the pure asphalt.

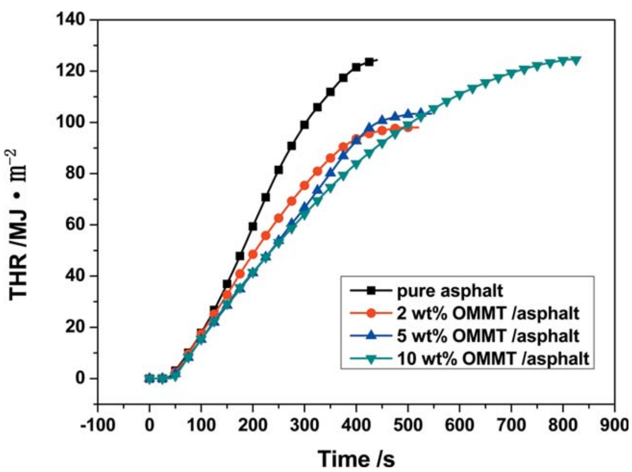

Figure 6. THR curves of the CONE tests for the pure asphalt and OMMT-modified asphalts with different contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The smoke production could also be examined by the CONE test on the basis of the theory of the attenuation of a beam of light by suspended aerosol particulates. The amount of smoke was measured during all of the duration of the test, and the TSR was calculated as the ratio of the total smoke production to the exposed surface area. Figure 7 shows that addition of 2 wt % OMMT greatly reduced the value of TSR; this suggested that OMMT had a considerable effect on the smoke

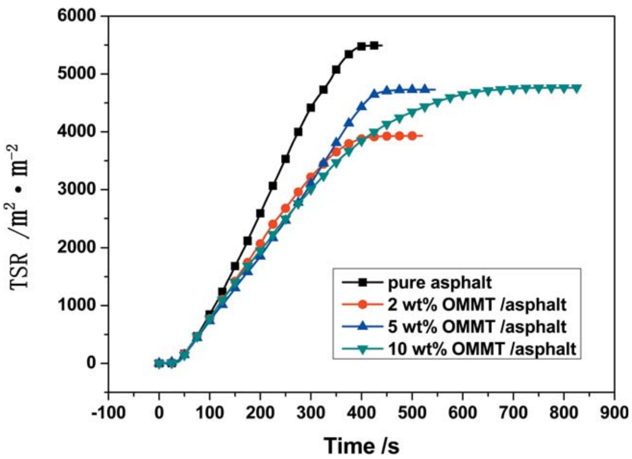

Figure 7. TSR curves of the CONE tests for the pure asphalt and OMMT-modified asphalts with different contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

suppression. However, there was almost no difference in TSR for asphalts modified with different contents of OMMT in the early stage of the combustion process; and the smoke production amounts of 5 and 10 wt % OMMT-modified asphalts were larger than that of the 2 wt % sample.

Possible Mechanism for the Effect of OMMT on the Flame Retardancy. With the results of both the LOI and CONE tests taken into account, we reached the conclusion that the incorporation of a relatively small content of OMMT dramatically improved the flame-retardant properties of the asphalt, for instance, by increasing the LOI; lowering the HRR, pkHRR, $MHRR_{360}$, THR, and TSR; and prolonging TTI. However, further increases in the OMMT content did not obviously improve the flame retardancy and even resulted in performance degradation, especially when the OMMT content was over 10 wt %.

The thermogravimetry (TG) and derivative thermogravimetry (DTG) curves of OMMT are shown in Figure 1. The mass loss of 15.7 wt %, which took place around 325°C, suggested that the content of the organic modification agent, octadecyl ammonium salt, was about 15 wt %. The combustibility of the

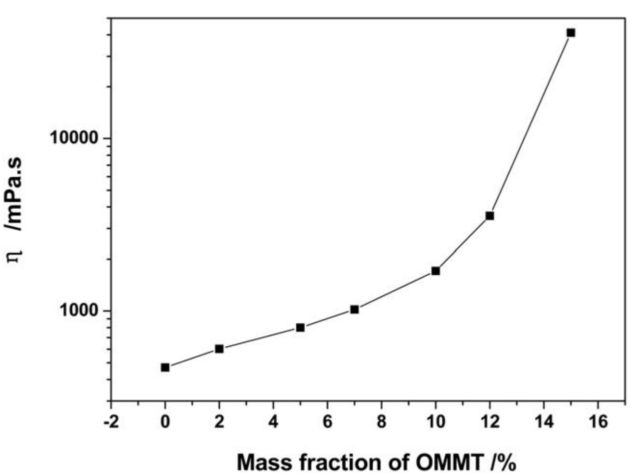

Figure 8. Curve of the asphalt viscosity (η) versus the OMMT content.

octadecyl ammonium salt was similar to the light component in the asphalt because both of them had a long hydrocarbon chain structure; therefore, the octadecyl ammonium salt should have exhibited poor flame retardancy compared to the pure asphalt. We assumed that OMMT should have had dual effects on the flame retardancy of the asphalts. On the one hand, the intercalated structure formed by the nanodispersed OMMT effectively hindered the penetration of oxygen inside the asphalt and retarded the burning course of asphalts. This positive effect was similar to that in clay/polymer nanocomposites.[19] On the other hand, the octadecyl ammonium salt contained in OMMT worsened the flame-retardant properties of the asphalts because of its high combustibility. When the OMMT content was relatively small, the positive effect was dominant; therefore, the flame-retardant properties could be improved dramatically. With increasing OMMT content, the octadecyl ammonium salt content became considerable, and as a result, the negative effect turned out to be obvious and offset the positive effect. This might be main reason why the modified asphalt with a high content of OMMT showed worse flame-retardant properties than that with a low content of OMMT.

Effect of OMMT on the Physical Properties of Asphalt

Effect of OMMT on the Viscosity of Asphalt. The viscosity reflects the flow-resistance ability of asphalt under a high temperature. The higher the viscosity is, the better the rutting-resistance performance asphalts have. So, the viscosity is an important index for evaluating the high-temperature performance of asphalts. Figure 8 displays the influence of the OMMT content on the viscosity of the modified asphalt. We observed that with increasing OMMT content, the viscosity of the OMMT-modified asphalt increased; this suggested improvement in the rutting resistance and high-temperature performances of the asphalts. This result was ascribed to the fact that the movement of the asphalt molecule was confined by the intercalated layers of OMMT. According to Strategic Highway Research Program specifications, the viscosity should be below 3 Pa s at 135°C. However, the viscosity values of the 12 and 15 wt % OMMT-modified asphalt were higher than 3 Pa s; this means that it could not be used in asphalt concrete. This sharp increase in its viscosity could be ascribed to the strong filler–network structure formed when the OMMT content was high (shown in Figure 3).

Effect of OMMT on the Other Physical Properties of Asphalt. The softening point is usually used to evaluate high-temperature resistance to deformation and the high-temperature stability of the asphalt. The penetration of asphalt is a representation of asphalt consistency and reflects its rheological properties. The ductility is usually used to evaluate the low-temperature anticracking performance of asphalt. Table III shows the test results of the softening point, penetration, and ductility of various modified asphalt samples. With increasing OMMT content, the softening point increased, and the penetration obviously decreased. These phenomena could be explained by the formation of the intercalated structure in the OMMT-modified asphalt. These OMMT layers with high aspect ratios greatly obstructed the movement of the asphalt molecular chains. These results also indicated that OMMT improved the

Table III. Physical Properties of the OMMT/Asphalt Composites

Sample	Softening point (°C)	Pen_{25} (dmm)	Ductility at 5°C (cm)	PI
Pure asphalt	46.0	79	7.5	−1.170
2 wt % OMMT/asphalt	50.4	70	6.5	−0.274
5 wt % OMMT/asphalt	51.9	68	7.0	0.028
7 wt % OMMT/asphalt	53.2	61	6.9	0.056
10 wt % OMMT/asphalt	57.7	42	4.6	0.141
12 wt % OMMT/asphalt	71.3	40	3.8	2.473
15 wt % OMMT/asphalt	87.0	36	2.8	4.382

high-temperature stability of the asphalt. The ductility of the OMMT-modified asphalt at 5°C changed slightly compared to that of the pure asphalt when the OMMT content was not over 7 wt %. With further increasing OMMT content, the cracks formed by interfacial debonding quickly converged; this resulted in early fracture; therefore, the ductility dropped down sharply. The temperature sensitivity of the OMMT-modified asphalt was calculated in terms of the penetration index (PI), also shown in Table III. An approach related to PI calculation can be found in the Shell Bitumen Handbook according to the following equation:[26]

$$PI=\frac{1952\text{-}500\times\log(Pen_{25})-20\times SP}{50\log\times(Pen_{25})\text{-}SP\text{-}120} \quad (2)$$

where Pen_{25} is the penetration at 25°C in tenths of millimeters and SP is the softening point temperature of OMMT-modified asphalt (°C). We found that the PI values increased with increasing OMMT content. Because the higher PI values indicated a lower thermal sensitivity, this result revealed that the OMMT-modified asphalt was more resistant to low-temperature cracking and high-temperature rutting.

CONCLUSIONS

XRD analyses indicated that asphalt molecules could intercalate into the interlayer space of OMMT and form an intercalated structure after melt blending at high temperature. When the OMMT content increased from 2 to 15 wt %, the interlayer space of the intercalated structure decreased slightly. RPA implied the formation of a filler–network structure in the OMMT-modified asphalt. A concentration of 15 wt % of OMMT was too much to achieve a good dispersion of OMMT in the asphalt.

LOI tests showed that the addition of OMMT concentrations of lower than 7 wt % increased the LOI of the asphalt. The LOI of the asphalt containing 7 wt % OMMT reached 31.1%. The CONE tests also showed that the incorporation of small amounts of OMMT obviously decreased the values of HRR, pkHRR, THR, TSR, and $MHRR_{360}$ and prolonged TTI. However, further increases in the OMMT content could not obviously improve the flame retardancy and even resulted in performance degradation, especially when the OMMT content was over 10 wt %; this was possibly due to the introduction of a considerable amount of a flammable organic modifier.

As the OMMT content increased, the viscosity, softening point, and PI value of the OMMT-modified asphalt increased, but the penetration decreased; this indicated improved high-temperature stability and lowered temperature sensitivity. When the OMMT content was not over 7 wt %, the ductility at 5°C of the modified asphalt decreased slightly compared with that of the pure asphalt.

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