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Research Article

Preparation of Flame Retardant Modified with Titanate for Asphalt Binder

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Improving the compatibility between flame retardant and asphalt is a difficult task due to the complex nature of the materials. This study explores a low dosage compound flame retardant and seeks to improve the compatibility between flame retardants and asphalt. An orthogonal experiment was designed taking magnesium hydroxide, ammonium polyphosphate, and melamine as factors. The oil absorption and activation index were tested to determine the effect of titanate on the flame retardant additive. The pavement performance test was conducted to evaluate the effect of the flame retardant additive. Oxygen index test was conducted to confirm the effect of flame retardant on flame ability of asphalt binder. The results of this study showed that the new composite flame retardant is more effective in improving the compatibility between flame retardant and asphalt and reducing the limiting oxygen index of asphalt binder tested in this study.

1. Introduction

Asphalt is a complex mixture of organic molecules that vary in chemical compositions and molecular weights [1–3]. Owing to the asphalt binder chemical constitution, however, the material is quite flammable and produces smoke and poisonous gases while burning [4]. Due to these properties, it is dangerous to use asphalt binder in road tunnel pavements, gas stations, and so forth [5–7]. An expanded growth of asphalt application concurrent with the proliferation of safety standards being set by government and private agencies has indicated that it is of primary importance to reduce the flammability of asphalt materials [8].

Generally, five burning processes are considered to be involved in asphalt flammability: heating, decomposition, ignition, combustion, and propagation. Flame retardation can be accomplished by the disruption of the burning process at one or more stages so that the process is terminated within an acceptable period of time, preferably before ignition actually occurs [8]. Asphalt binders with excellent flame retardation capability have been prepared successfully by adding various flame retardants such as antimony trioxide, decabromodiphenyl ether, aluminum trihydroxide, and zinc borate. Different flame retardants exhibit different mechanisms of flame resistance [9–11]. So far, halogen-based flame retardants have been extensively employed for this purpose, but the halogen-containing materials easily released a good deal of poisonous smoke and corrosive fumes in their burning process [12–14]. As consciousness increases in environmental protection and technology advances in flame retardants, the development trend of flame retardants will be halogen-free, nontoxic, and smoke suppressing [15, 16]. Most halogen-free formulations are based on inorganic fillers such as metallic hydroxide and expandable graphite [17]. In recent years, magnesium hydroxide has become one of the most popular replacements for halogen-based flame retardants and has been widely used in polymer materials [18]. However, the disadvantages of inorganic fire-retardant agents such as low fire-retardant efficiency, large adding amount, strong polarity, and poor compatibility with asphalt limit its applications

[19, 20]. Furthermore, it still has a potential threat for pavement performance at larger concentration of around 20% by weight. Therefore, it is necessary to develop a low dosage of flame retardant, to create equilibrium between flame retardant properties and pavement performance.

Since magnesium hydroxide, ammonium polyphosphate, and other materials are inorganic fillers, which have inert and hydrophilic nature, they are of poor compatibility with asphalt. Coarse particles lack the reinforcement. Finer particles often have poor dispersion, which can cause particles to become aggregates. Therefore, it is necessary to use surface treatment of inorganic fillers in order to overcome the defect itself and receive the new physical and chemical properties in the surface of the particle. In recent years, titanate coupling agent is used to modify the inorganic fillers [21, 22] because the titanate coupling agent has a unique structure which has a good coupling effect for the polymer and filler. Therefore, it can improve the dispensability of the flame retardant in asphalt and improve flame retardancy.

The objective of this study is to develop a low dosage compound flame retardant and improve the compatibility between flame retardants and asphalt. An orthogonal experiment was designed taking magnesium hydroxide, ammonium polyphosphate, and melamine as factors. The oil absorption and activation indexes were tested to determine the effect of titanate on the flame retardant additive. The pavement performance test was conducted to evaluate the effect of the flame retardant additive. Oxygen index test was conducted to confirm the effect of flame retardant on flame ability of asphalt binder.

2. Materials and Test Program

2.1. Materials. An asphalt binder was used in this study. It was from Kelamayi crude source. Table 1 summarizes the properties of the binder in this study.

Three main ingredients of the flame retardants used in this paper include magnesium hydroxide, melamine, and ammonium polyphosphate. Magnesium hydroxide is a new type of flame retardant filler. It can reduce the surface temperature of the asphalt while burning because it releases water and can absorb large quantities of latent heat when it is heated, which is helpful in inhibiting the decomposition of the polymer and cooling the generated combustible gas. Ammonium polyphosphate (APP) has the role of the acid source, which can form an intumescent flame retardant system and have a better synergistic flame retardant effect with $Mg(OH)_2$. Melamine itself has flame retardance whose decomposition temperature is over 300°C, which can be dissolved in the asphalt. As described above, magnesium hydroxide, ammonium polyphosphate, and melamine can be treated by surface polarity activation using a titanate coupling agent, which can improve flame retardant's lipophilicity and dispersibility. And their physical properties are shown in Tables 2 and 3.

2.2. Preparation of Flame Retardant. Magnesium hydroxide, ammonium polyphosphate, and melamine were mixed together and dried in the oven to reduce their surface water after they were weighed accurately using an electronic balance. We can name the composite flame retardant as FRA. The titanate coupling agent was added in the composite flame retardant. Then they were mixed well with a stirrer and dried in the oven. A new flame retardant was obtained. We can name it as FRA-Ti. Their quality and proportion were determined by orthogonal table, respectively. Table 4 shows the factors and levels in orthogonal test. Table 5 shows the factors combination in an orthogonal test.

2.3. Preparation of Asphalt Binder Modified with Flame Retardant. The asphalt was modified by adding the different FRA-Ti into asphalt binder and mixing with the stirrer. The specific preparation process and steps are as follows. 1000 g asphalt was heated at about 140°C after it was dehydrated at 105°C. And 50 g FRA-Ti was added into the asphalt binder and stirred about 30 min at 500 r/min.

2.4. Pavement Performance. The pavement performance test was conducted to evaluate the effect of the flame retardant additive including the softening point, penetration, ductility, and elastic recovery in accordance with the test requirements of Technical Specification for Construction of Highway Asphalt Pavements (JTG F40—2004).

2.5. Activation Index. Modified activation index of flame retardancy was measured by the following method: a cylinder volume of 20 mL water was added to a separatory funnel, along with 5 g of the modified flame retardant, 1 min shaking up and down 120 times, and allowed to stand over 1 h, and then opening the stopcock deposition on the bottom of the sample discharge, drying, weighing, with the original mass weighed (5 g) minus the mass of the sample deposition indicates the floating portion of the original mass. Activation index (AI) is calculated as follows:

$$\mathrm{AI} = \frac{(m_0 - m_1)}{m_0} \times 100\%, \quad (1)$$

where m_0 is original mass weighed (5 g) and m_1 is the mass after activation.

2.6. Oil Absorption. Oil absorption rate was measured by titrating fat dioctyl phthalate (DOP) in the flame retardant samples. In the process of titration, the flame retardant should be constantly turning. It reaches the titration end point when flame retardant resembles "dough." And also note the before and after titration DOP quality. The oil absorption rate (OAR) is defined in

$$\mathrm{OAR} = \frac{M_{\mathrm{DOP}}}{M_{\mathrm{FRA}}} \times 100\%, \quad (2)$$

where M_{DOP} is used mass of DOP and M_{FRA} is the original mass of flame retardant.

2.7. Limiting Oxygen Index. Flame retardancy of modified asphalt binder was evaluated by the limiting oxygen index

Table 1: Properties of virgin binders.

Test items	Penetration (100 g, 5 s, 25°C)/0.1 mm	Ductility (5°C)/cm	Softening point/°C	Residue after RTFOT		
				Mass loss	Penetration ratio of 25°C/%	Ductility (5°C)/cm
KLMY-90	98	29.7	48	−0.188	80	28.8

Table 2: Physical properties of $Mg(OH)_2$, APP, and MEL.

	$Mg(OH)_2$	APP	Melamine
Outward appearance	White powder	White powder and good dispersion	White single crystals
Density	2.388 g/cm^3	1.732 mg/cm^3	1.572 mg/cm^3
Initial decomposition temperature	>300°C	>250°C	>300°C

Table 3: Physical properties of titanate coupling agent.

Test items	Outward appearance	Molecular	Molecular weight	Content	Density	Benzene dissolution test
Titanate	Buff oily liquid	$C_{16}H_{36}O_4Ti$	340.36	≥98%	0.999–1.003	Qualified

Table 4: The factors and level in orthogonal test.

Level	(A) $Mg(OH)_2$ (g)	(B) APP (g)	(C) Melamine (g)	(D) Titanate (%)
1	25	33	15	5
2	20	35	17.5	5.5
3	15	38	20	6

Table 5: The design of orthogonal test.

	A	B	C	D	Orthogonal combination
1	1	1	1	1	$A_1B_1C_1D_1$
2	1	2	2	2	$A_1B_2C_2D_2$
3	1	3	3	3	$A_1B_3C_3D_3$
4	2	1	2	3	$A_2B_1C_2D_3$
5	2	2	3	1	$A_2B_2C_3D_1$
6	2	3	1	2	$A_2B_3C_1D_2$
7	3	1	3	2	$A_3B_1C_3D_2$
8	3	2	1	3	$A_3B_2C_1D_3$
9	3	3	2	1	$A_3B_3C_2D_1$

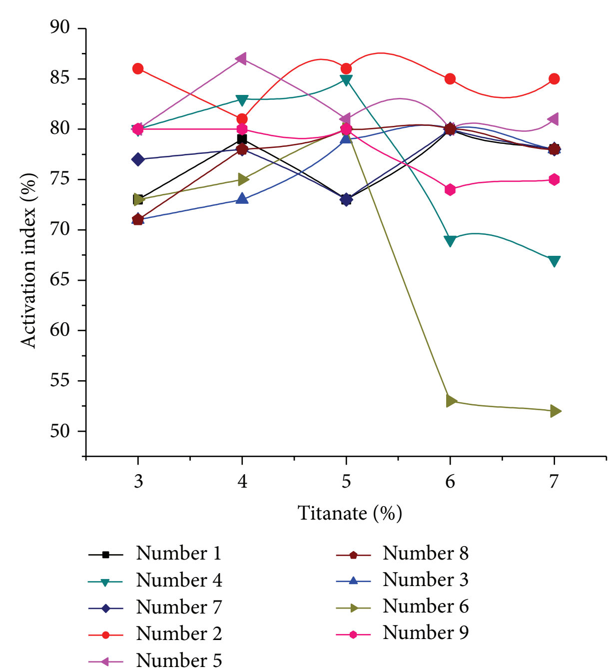


Figure 1: Oil absorption of FRA modified by titanate.

(LOI, HC-2, Jiangning Instruments Co., Ltd., China). Limiting oxygen index methods are widely used to measure the flammability of polymers and to investigate the effectiveness of flame retardants. Flame retardancy was assessed by the LOI according to ASTM D-2863. For testing, the top of the sample was ignited by a gas flame, which was stopped once ignition has occurred. Then, the lowest oxygen concentration in a flowing mixture of nitrogen and oxygen, which just supports sustained burning, can be determined. The effectiveness of flame retardants is measured by the changes in the critical oxygen concentration. The LOI is defined in

$$\mathrm{LOI} = \frac{\Phi_{cr}(O_2)}{\Phi_{cr}(O_2) + \Phi(N_2)}, \quad (3)$$

where $\Phi_{cr}(O_2)$ and $\Phi(N_2)$ are the minimum oxygen concentration and the relevant nitrogen concentration in the inflow gases, respectively.

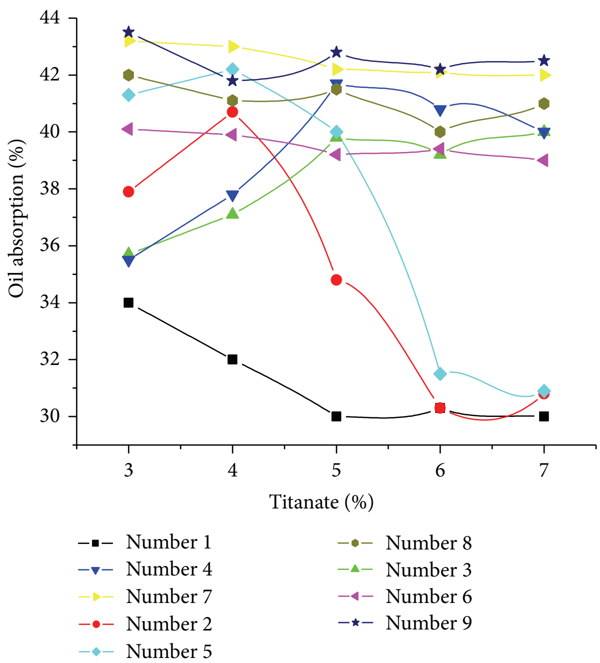


Figure 2: Activation index of FRA modified by titanate.

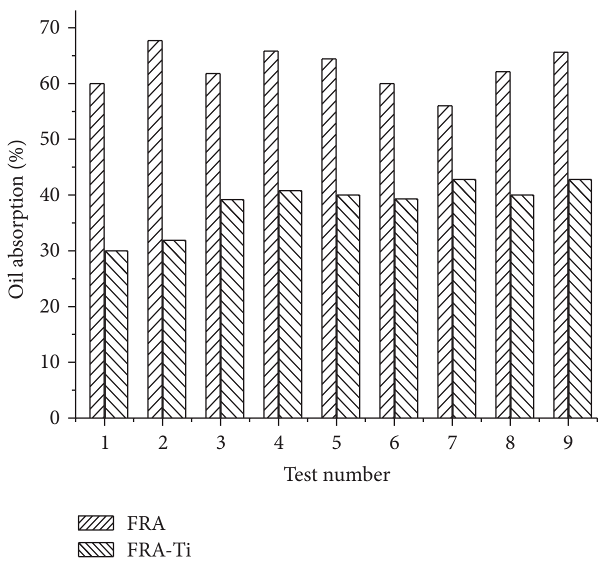


Figure 3: Oil absorption of FRA and FRA-Ti.

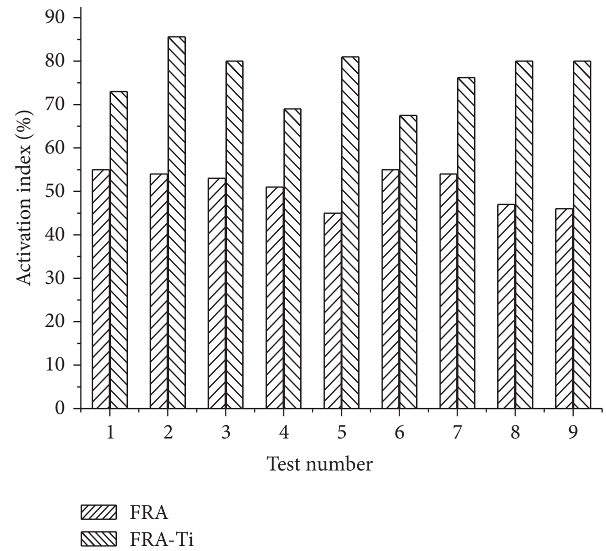


Figure 4: Activation index of FRA and FRA-Ti.

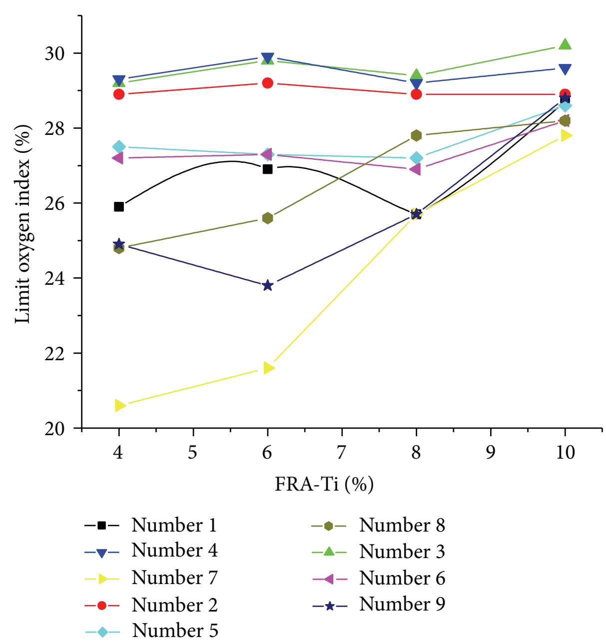


Figure 5: Limiting oxygen index (LOI) of asphalt with FRA-Ti.

3. Results and Discussions

3.1. Effect of Titanate on the Oil Absorption and Activation Index. After the titanate coupling agent was added, the oil absorption and activation index of nine FRA were measured and results are illustrated in Figures 1 and 2, respectively. A general trend found that oil absorption of FRA with titanate was stable 40–44% except number 1, number 2, and number 5. After titanate amount increasing to 6%, the oil absorption rate reached a stable value. The general trend of activation index is similar to the findings for oil absorption as shown in Figure 1. After the titanate amount was increased to 5%, their activation index reached a stable value except number 4, number 5, and number 6. The main reason is thought to be that FRA particle surface is gradually covered by coupling molecules with the increase of the amount of titanate, which lead the surface polarity to a gradual transition from hydrophilic to lipophilic, decreased oil absorption, and instable activation index. When titanate was added to the optimal dosage, the surfaces of each FRA particle were covered by a titanate coupling agent. After more than the optimum amount, the excess coupling is only dispersed in the organic phases, which have little effect on surface modification for FRA [23]. It indicated that the optimal dosage of titanate is 6% considering the oil absorption and activation index of nine FRA.

The results of oil absorption and activation index of nine FRA with and without titanate coupling agent are showed in Figures 3 and 4, respectively. The general trend found that oil absorption rate decreased and activation index increased for FRA modified by titanate.

3.2. Effect of Flame Retardant on the Limiting Oxygen Index. Limiting oxygen index estimates the flame residence. It is defined as the lowest O_2 concentration to maintain burning in mixed gas with N_2 and O_2. The O_2 concentration in air is 21%. When LOI $\leq$ 21%, this material is easily burned; when LOI is between 21% and 27%, this material can burn; and when LOI $\geq$ 27%, this material should be autoextinguished. The LOI of flame-retarded asphalt which was above 23% was put forward by Hui-qiang and Shi-zhou [24]. Figure 5 shows the LOI result of nine FRA at different proportion. The general trend found that the LOI of asphalt with FRA-Ti is 3% larger than that of virgin asphalt (21.3%). Though LOI of No7 is not changed obviously at the 4% and 6% FRA-Ti, it increased and reached the same level when 8% FRA-Ti was added into asphalt. We can conclude that the minimum FRA-Ti needed to improve the limiting oxygen index of asphalt is 8%. In addition, the LOI of number 3 was the highest, which achieved 29.8% and belonged to the autoextinguished materials. It indicated that flame retardant is helpful in improving the flame retardant properties of asphalt.

3.3. Effect of Flame Retardant on the Pavement Performance of Asphalt Binder. Figure 6 shows the pavement performance indicators of asphalt binder with different dosage flame retardant. A general trend of softening point increase with increased dosage of FRA-Ti is observed, and the penetration, ductility, and elastic recovery become smaller. It can be concluded that the effect of flame retardant on pavement performance of asphalt is significant. The appropriate flame retardant and its dosage in the application process should be selected in order to guarantee the pavement performance of asphalt binder.

3.4. Determining the Formulations of Flame Retardant. As can be seen from the above analysis, the optimal dosage is 6% for nine FRA-Ti. The pavement performance and LOI results of asphalt binder with nine FRA-Ti are shown in Table 6.

As mentioned above, the flame retardant not only must ensure the good flame retardant performance in asphalt but also should not significantly affect its pavement performance. Therefore, the several important factors should be considered when selecting the best formula for flame retardants. Firstly, it should be of consideration that the oil absorbing and activation index of nine FRA-Ti meet the basic requirements for flame retardant modification. In other words, oil absorption rate value should be small and activation index value is larger. Secondly, the pavement performance and LOI are important factors. In order to analyze the effect of every factor on the asphalts, the weights were calculated by the method of multi-index comprehensive score. It carries on comprehensive score calculation and comprehensive evaluation to merits based on confirming the index weight and index data normalization. A multi-index synthetic evolution model of weighted summation and weighted for components combination was selected [25] where Index is the test result of the index, $\mathrm{Min}_{\mathrm{index}}$ is minimum of the index and $\mathrm{Max}_{\mathrm{index}}$ is maximum of the index. Index membership is defined in

$$\text{Membership} = \frac{\text{Index} - \mathrm{Min}_{\mathrm{index}}}{\mathrm{Max}_{\mathrm{index}} - \mathrm{Min}_{\mathrm{index}}}. \quad (4)$$

The index membership of pavement performance, LOI, oil absorbing, and activation index of nine asphalts with FRA-Ti are shown in Table 7. It is evident that composite score of number 3 FRA-Ti was the highest. And it is a best recipe of flame retardant for asphalt binder. In addition, the sort of primary and secondary effects on the overall index factors is $Mg(OH)_2$, melamine, APP, and titanate.

4. Conclusion

In this paper, a self-developed low dosage compound flame retardant has been prepared using orthogonal tests including four factors and three levels. The compound flame retardant was surface modified by titanate. The oil absorption and activation index were tested to evaluate the effect of titanate on the flame retardant. A series of asphalt binder tests were carried out to confirm the effect of flame retardant on the pavement performance. Oxygen index was tested to determine the effect of flame retardant on flammability of asphalt binder. From these test results, the following conclusions were drawn for the materials used in this study.

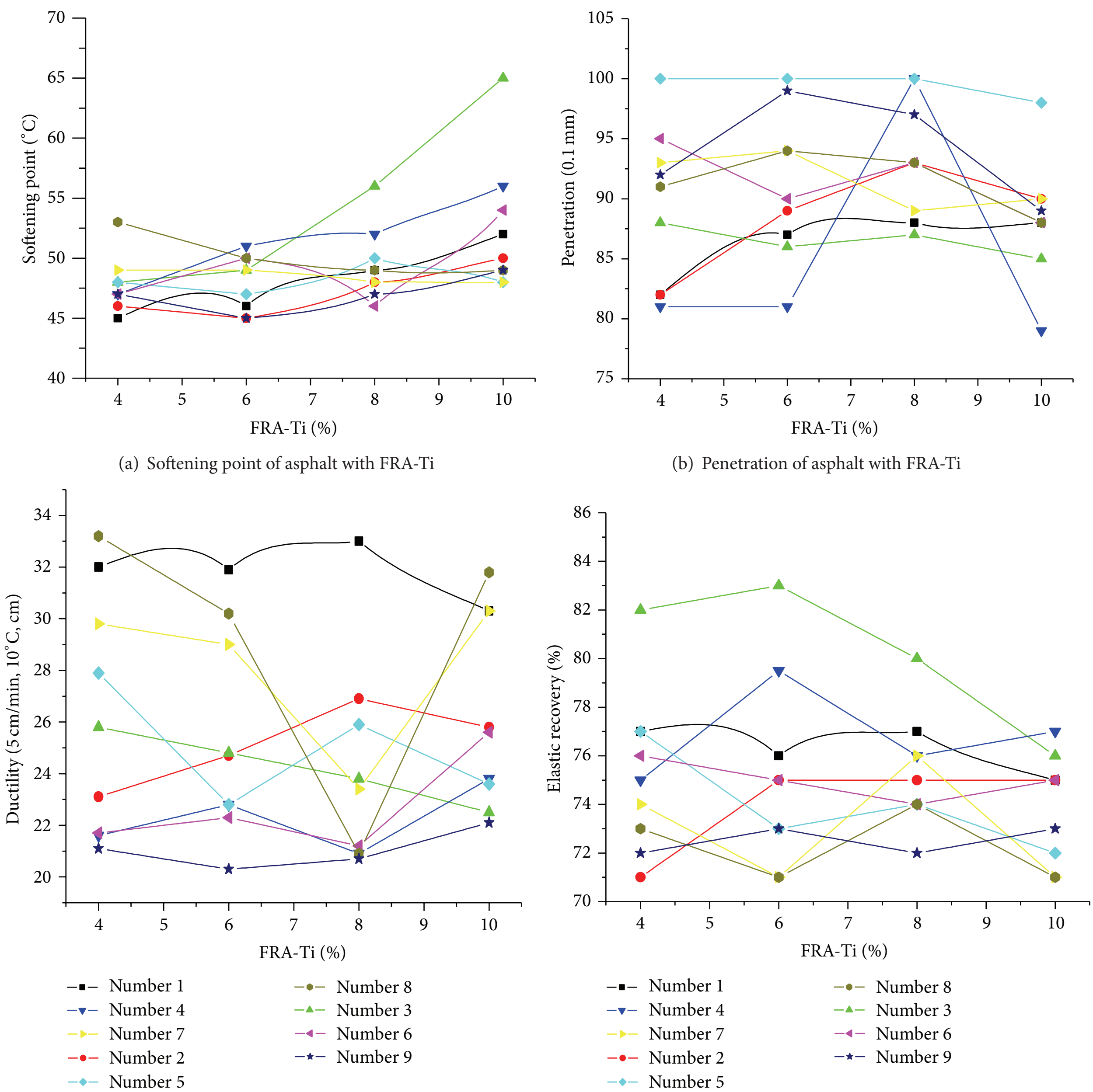
(1) The impact of titanate activation index and oil absorbing for flame retardants is significant. The optimal dosage of titanate is 6% for compound flame retardant considering the oil absorption and activation index.

(2) The compound flame retardant can significantly improve limiting oxygen index of asphalt binder. And the maximum increase is up to 8%.

(3) The effect of flame retardant on pavement performance for asphalt binder is significant. One should select the appropriate flame retardant and its dosage in the application process in order to guarantee the pavement performance of asphalt binder.

(4) The addition of flame retardant, including magnesium hydroxide, ammonium polyphosphate, and melamine, can affect pavement performance and LOI differently. The best recipe of flame retardant for asphalt binder is 30% magnesium hydroxide, 46% ammonium polyphosphate, and 24% melamine.

(5) It is recommended to conduct another study to evaluate pavement performance and burning behavior of asphalt mixture using the flame retardant modified with titanate. Also, further study with many other flame retardant and surface modifications is needed to generalize these findings.



(a) Softening point of asphalt with FRA-Ti

(b) Penetration of asphalt with FRA-Ti

(c) Ductility of asphalt with FRA-Ti

(d) Elastic recovery of asphalt with FRA-Ti

Figure 6: Pavement performance of asphalt with FRA-Ti.

Table 6: Test results of asphalt with FRA-Ti.

	Softening point (°C)	Penetration (0.1 mm)	Ductility (5 cm/min, 10°C, cm)	Elastic recovery (%)	LOI (%)
Number 1	46	87	31.9	76	26.9
Number 2	45	89	24.7	75	29.2
Number 3	48	92	24.5	78	29.8
Number 4	51	81	22.8	79.5	29.9
Number 5	47	100	22.8	73	27.3
Number 6	50	90	22.3	75	27.3
Number 7	49	94	29	71	21.6
Number 8	50	94	30.2	71	25.6
Number 9	45	99	20.3	73	23.8

Table 7: The index membership and composite score of FRA-Ti.

	A	B	C	D	Index membership: Oil absorbing	Activation index	Softening point	Penetration	Ductility	Elastic recovery	LOI	Composite score
Number 1	1	1	1	1	0	0.3	0.22	0.31	1	0.42	0.64	0.61
Number 2	1	2	2	2	0.15	1	0	0.41	0.38	0.33	0.92	0.64
Number 3	1	3	3	3	0.72	0.69	0.66	0.26	0.39	1	0.99	0.77
Number 4	2	1	2	3	0.84	0.08	0.99	0	0.22	0.71	1	0.59
Number 5	2	2	3	1	0.78	0.75	0.42	1	0.22	0.17	0.69	0.62
Number 6	2	3	1	2	0.73	0	0.8	0.49	0.17	0.33	0.69	0.27
Number 7	3	1	3	2	1	0.48	0.62	0.67	0.75	0	0	0.55
Number 8	3	2	1	3	0.78	0.69	0.77	0.69	0.85	0	0.48	0.49
Number 9	3	3	2	1	1	0.69	0.02	0.93	0	0.17	0.27	0.29
K_1	2.01	1.74	1.36	1.51								
K_2	1.47	1.74	1.51	1.45								
K_3	1.32	1.32	1.93	1.84								
k_1	0.67	0.58	0.45	0.5								
k_2	0.49	0.58	0.5	0.48								
k_3	0.44	0.44	0.64	0.61								
R	0.23	0.14	0.19	0.13								
Sort	ACBD											
Best recipe	$A_1C_3B_3D_3$											

Note. K_1, K_2, and K_3 are sum of the index membership composite score of every sample at every factor and level, respectively. k_1, k_2, and k_3 are the average value of the index membership composite score of every sample at every factor and level, respectively. R is the range of the index membership composite score of every sample at every factor and level, respectively.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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References

[1] P. K. Das, N. Kringos, and B. Birgisson, "Microscale investigation of thin film surface aging of bitumen," *Journal of Microscopy*, vol. 254, no. 2, pp. 95–107, 2014.

[2] G. Wen, Y. Zhang, Y. Zhang, K. Sun, and Y. Fan, "Rheological characterization of storage-stable SBS-modified asphalts," *Polymer Testing*, vol. 21, no. 3, pp. 295–302, 2002.

[3] H. Yao, Z. You, L. Li et al., "Rheological properties and chemical analysis of nanoclay and carbon microfiber modified asphalt with Fourier transform infrared spectroscopy," *Construction and Building Materials*, vol. 38, pp. 327–337, 2013.

[4] S. Wu, L. Mo, P. Cong, J. Yu, and X. Luo, "Flammability and rheological behavior of mixed flame retardant modified asphalt binders," *Fuel*, vol. 87, no. 1, pp. 120–124, 2008.

[5] Y. Qun and Z. Y. Guo, "Mixture design of fire-retarded OGFC in road tunnel," *Road Materials and Pavement Design*, vol. 6, no. 2, pp. 255–268, 2005.

[6] A. Leitner, "The fire catastrophe in the Tauern Tunnel: experience and conclusions for the Austrian guidelines," *Tunnelling and Underground Space Technology*, vol. 16, no. 3, pp. 217–223, 2001.

[7] A. Lönnermark and H. Ingason, "Gas temperatures in heavy goods vehicle fires in tunnels," *Fire Safety Journal*, vol. 40, no. 6, pp. 506–527, 2005.

[8] S. Wu, P. Cong, J. Yu, X. Luo, and L. Mo, "Experimental investigation of related properties of asphalt binders containing various flame retardants," *Fuel*, vol. 85, no. 9, pp. 1298–1304, 2006.

[9] "Fire retardant asphalt composition. Add polymer," Patent US 5026747, 1991.

[10] H. Demir, E. Arkiş, D. Balköse, and S. Ülkü, "Synergistic effect of natural zeolites on flame retardant additives," *Polymer Degradation and Stability*, vol. 89, no. 3, pp. 478–483, 2005.

[11] A. Toldy, N. Tóth, P. Anna, and G. Marosi, "Synthesis of phosphorus-based flame retardant systems and their use in an epoxy resin," *Polymer Degradation and Stability*, vol. 91, no. 3, pp. 585–592, 2006.

[12] F. Laoutid, L. Ferry, J. M. Lopez-Cuesta, and A. Crespy, "Flame-retardant action of red phosphorus/magnesium oxide and red phosphorus/iron oxide compositions in recycled PET," *Fire and Materials*, vol. 30, no. 5, pp. 343–358, 2006.

[13] M. Sain, S. H. Park, F. Suhara, and S. Law, "Flame retardant and mechanical properties of natural fibre-PP composites containing magnesium hydroxide," *Polymer Degradation and Stability*, vol. 83, no. 2, pp. 363–367, 2004.

[14] J. M. Laza, E. Bilbao, M. T. Garay, J. L. Vilas, M. Rodríguez, and L. M. León, "Thermal properties and fire behaviour of materials produced from curing mixed epoxy and phenolic resins," *Fire and Materials*, vol. 32, no. 5, pp. 281–292, 2008.

[15] X. Chen, J. Yu, and S. Guo, "Structure and properties of polypropylene composites filled with magnesium hydroxide," *Journal of Applied Polymer Science*, vol. 102, no. 5, pp. 4943–4951, 2006.

[16] S. A. A. Ramazani, A. Rahimi, M. Frounchi, and S. Radman, "Investigation of flame retardancy and physical-mechanical properties of zinc borate and aluminum hydroxide propylene composites," *Materials and Design*, vol. 29, no. 5, pp. 1051–1056, 2008.

[17] N. Huang and J. Wang, "A TGA-FTIR study on the effect of $CaCO_3$ on the thermal degradation of EBA copolymer," *Journal of Analytical and Applied Pyrolysis*, vol. 84, no. 2, pp. 124–130, 2009.

[18] T. Xu and X. Huang, "Pyrolysis properties and kinetic model of an asphalt binder containing a flame retardant," *Journal of Applied Polymer Science*, vol. 119, no. 5, pp. 2661–2665, 2011.

[19] H.-Q. Chen and P.-W. Hao, "Development of the modified fire-retardant asphalt and performance evaluation on its mixture," *Journal of Wuhan University of Technology*, vol. 30, no. 12, pp. 58–62, 2008.

[20] S.-Y. Lu and I. Hamerton, "Recent developments in the chemistry of halogen-free flame retardant polymers," *Progress in Polymer Science*, vol. 27, no. 8, pp. 1661–1712, 2002.

[21] G. Chunsuo and C. Chengzhi, "Study on surface modification of inorganic fillers with titanate coupling agent," *Technology & Development of Chemical Industry*, vol. 36, no. 9, pp. 4–7, 2007.

[22] M. Zhiling, F. congran, and M. Xin, "Coupling function of titanate coupling agent in PP/IFR composites," *China Plastics*, vol. 25, no. 10, pp. 77–80, 2011.

[23] H.-Q. Chen and P.-W. Hao, "Research on the surface modification of fire-retardant asphalt with titanate coupling agent," *Journal of Wuhan University of Technology*, vol. 31, no. 17, pp. 66–69, 2009.

[24] C. Hui-qiang and C. Shi-zhou, "Research on asphalt flame re-tardant modification technology," *Journal of Highway and Transportation Research and Development*, no. 2, pp. 19–20, 2003.

[25] B. Li, S. Han, O.-M. Xu, and X.-Q. Teng, "Evaluation of asphalt pavement performance based on principal component analysis," *Journal of Chang'an University*, vol. 29, no. 3, pp. 15–18, 2009.