



Characterization of zirconium tungstate filler and performance investigation on asphalt mastic made with zirconium tungstate filler



Junyan Yi^{a,b,*}, Yixiang Cao^c, Decheng Feng^a, Yudong Huang^b

^aSchool of Transportation Science and Engineering, Harbin Institute of Technology, Harbin 150090, China

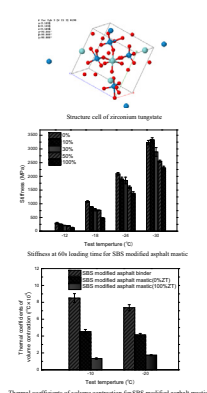
^bSchool of Chemical Engineering and Technology, Harbin Institute of Technology, Harbin 150090, China

^cGuangdong Provincial Academy of Building Research Group Co., Ltd., Guangzhou 510500, China

HIGHLIGHTS

- A new-developed material-ZrW₂O₈ with negative thermal expansion properties has been used as a substitute of mineral filler.
- The physical properties of ZrW₂O₈ fillers and mineral fillers are investigated.
- The performances of asphalt mastic made with ZrW₂O₈ fillers are studied.

GRAPHICAL ABSTRACT



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ABSTRACT

Thermal cracking is a widespread distress for asphalt pavement in cold regions. The main reason causing thermal cracking is due to the contraction of asphalt concrete. Once thermally induced contraction stress exceeds the fracture resistance of the asphalt concrete, cracks start to develop. Comparing to aggregate, asphalt mastic is believed to have much larger thermal coefficient of contraction, therefore play a more significant role in determining the thermal cracking resistance of asphalt pavement. To modify the inherent thermal property of asphalt mastic, a new-developed material, zirconium tungstate (ZT), was utilized in this study. Different from normal material's heated expansion and cooled contraction properties, ZT has a negative thermal expansion property of heated contraction and cooled expansion. Being as a substitute for mineral filler, the physical properties of ZT fillers and performances of asphalt mastics made with ZT fillers were investigated. Results show that ZT can better absorb and adhere asphalt binder when comparing with mineral filler. The addition of ZT is able to decrease the thermal coefficients of volume contraction and creep stiffness, improve the adhesive bond between filler and asphalt binder and reduce the complex modulus of asphalt mastic.

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1. Introduction

Thermal cracking is one of the major causes of premature failures in asphalt pavements, and it also introduces many other distresses, such as moisture damage, potholes etc. It is normally

* Corresponding author at: School of Transportation Science and Engineering, Harbin Institute of Technology, Harbin 150090, China.

E-mail address: yijunyan@hit.edu.cn (J. Yi).

accepted that the mechanisms for thermal cracking can be divided into two kinds: single-event thermal cracking and thermal fatigue cracking. The former one refers to a very severe cold temperature or very fast cooling rate for pavement's environment. In that case, tensile stresses are generated in the asphalt layer due to its resistance to thermal contraction. Once it exceeds the tensile strength of asphalt concrete, thermal cracking generates at the surface of the asphalt layer. The colder temperature and faster cooling rate could result in the increase of quantity and width of thermal cracking. The thermal fatigue cracking is caused by the multiple cooling–warming cycles. Therefore this kind of cracking could also be found in the warmer region with higher daily temperature fluctuation.

Many researches had been made on the experimental techniques and evaluation methods for thermal cracking of asphalt pavement in the past forty years [1–5]. It was further concluded that the thermal cracking resistance of asphalt pavement was largely dependent on the pavement structure, environmental conditions and the physical and mechanical properties of the asphalt mixture (e.g. relaxation modulus and thermal coefficient of contraction) [6]. Among these influencing factors, the thermal coefficient of contraction of the asphalt mixture is one of the required properties in the prediction and evaluation of thermal cracking resistance of asphalt pavements [7]. The most famous empirical model, proposed by Haas and Phang at 1980s, which was used to predict the crack spacing of asphalt pavement, included the thermal coefficient of contraction of asphalt mixture as one of the most important predictive index [8].

As a typical composite material, asphalt mixture is blended with asphalt mastic (asphalt and filler) and aggregate. Comparing to asphalt mastic, which with time-dependent property, the aggregate has a more stable and constant physical and mechanical properties [9]. Therefore, the thermal coefficient of contraction of asphalt mastic was more determinative in the thermal fracture property of asphalt mixture, therefore could greatly affect the generation of thermal cracking [10]. Numerous researchers had conducted a lot of work on the binder modification to improve the cracking resistance of asphalt concrete [11,12].

As indicated before, the cooled contraction of asphalt concrete introduces the cumulative tensile stress, and thermal cracking generates once this stress exceeds the tensile strength of the asphalt concrete. As for the significant effect of thermal contraction property of asphalt mastic on the thermal property of the asphalt concrete, decreasing the thermal coefficient of contraction of asphalt mastic means the reduction of tensile stress and then thermal cracking potential in asphalt pavement. However, the thermal expansion–contraction is a natural property for most of existing materials, so is asphalt mastic. Thus most of present studies were focused on selecting better binder or increasing the effect of stone skeleton in asphalt mixture, to achieve a better cracking-resistance asphalt pavement.

Zirconium tungstate (ZT) is a new-developed material in recent years, whose molecular formula is ZrW_2O_8 . After Mary et al. reported the isotropic negative thermal expansion of this material at a large temperature range (0.3 K–1050 K), many researchers conducted a lot of studies on its special thermal property [13–16]. Specially the average negative thermal coefficients of contraction of ZT was reported to $-7.2 \times 10^{-6}/^{\circ}C$ [14]. Other researchers had presented the typical structural cell of ZT, and then characterize and explain the negative expansion mechanism of this material [15]. Some studies also tried to design composite materials which including ZT and other raw materials, to realize the production of new material with lower or even zero thermal expansion–contraction property. Actually, many kinds of composite materials with low thermal expansion property had been successfully produced in the last ten years, including

ZrW_2O_8 -Cu, ZrW_2O_8 -Al, ZrW_2O_8 -cement concrete, ZrW_2O_8 -epoxy and ZrW_2O_8 -CFRP etc. [17–22]. All these studies clearly indicated that the addition of ZT could reduce the thermal coefficients of contraction of composite materials.

In this study, in order to realize a better thermal property of asphalt-based material, ZT was introduced into the production of asphalt mastic by partially replacing the normal mineral filler. Then the physical properties of ZT filler and performance of asphalt mastic made with ZT were tested and compared with mastic made with mineral filler. The creep stiffness and volumetric thermal coefficient of expansion–contraction of different kinds of asphalt mastic at low temperatures were especially studied to investigate the low temperature performance.

2. Materials and experimental methods

2.1. Asphalt binders

Asphalt binder of 60/80 penetration grade (70# asphalt) was used as base material in this experiment. Styrene-butadiene-styrene modified asphalt (SBS asphalt) was also produced by mixing SBS modifier into the asphalt binder of 80/100 penetration grade. The basic properties of asphalt are shown in Table 1.

2.2. Fillers

Zirconium tungstate (ZT) utilized in this study was bought from one company in Yunnan Province of China. The cubic structure cell of ZT had been presented by Pryde in 1997 [15] and could be plotted in Fig. 1. This cubic structure was cited as Rigid Unit Modes (RUMs) and had a high volume expansion and contraction property [15]. In the structure, ZrO_6 with octahedron structure and WO_4 with tetrahedron structure are connected by the weak molecular bonding forces. Once temperature increases, ZrO_6 and WO_4 could have a coupling rotation which reduces the distance between Zr and W. The shortening of atoms' distance leads to the material's volume contraction, therefore forms the negative thermal expansion property of material. In this study, compositions of produced ZT were also measured and showed in Table 2.

Mineral filler made from limestone was used as the filler in asphalt mastic. The apparent density of filler is 2.61 g/cm^3 . The particle size distribution of filler was measured and compared to that of zirconium tungstate.

In order to replace the mineral filler, the properties of ZT were measured to analyze its feasibility as filler. Thus, physical property tests of fillers, including the apparent density, particle size distribution, specific surface area and pore size characterization, activity

Table 1
Properties of asphalt with 60/80 penetration grade and SBS modified asphalt.

Properties	70# asphalt	SBS asphalt
Penetration at 25 °C/0.1 mm	68	65.6
Penetration index (PI)	0.6	0.5
Softening point/°C	52.0	59.3
Ductility at 15 °C/cm	150	–
Ductility at 10 °C/cm	30	–
Ductility at 5 °C/cm	–	34
Density at 15 °C/(g/cm ³)	1.002	–
Wax content/%	1.5	–
Flashing point/°C	276	245
Segregation test during 48 h heated storage, difference of softening points for top and bottom part/°C	–	1.8
Retained penetration ratio after RTFOT at 25 °C/%	64	65
Retained ductility after RTFOT at 10 °C/cm	11	–
Retained ductility after RTFOT at 5 °C/cm	–	23

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