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Determining the effects of aging on halloysite nano-tube modified binders through the pull-off test method

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HIGHLIGHTS

- The HNT improved the adhesion force using stainless steel and granite substrates.
- The addition of the HNT decreases the effect of aging.
- The HNT may be considered as anti-aging and anti-stripping material.
- The pull-off test was able to evaluate the effects of aging on the adhesion force of binders.

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ABSTRACT

Aging changes the chemistry of bitumen and this adversely affects bitumen's adhesion with aggregate. For this reason, the use of pull-off test was proposed to evaluate bitumen's adhesion during different aging stages. In this study, the pull-off test method is used to evaluate the effects of aging on a 60/70 penetration grade bitumen and binders modified with 2 and 4% halloysite nano-tube (HNT). The short-term aging (STA) and long-term aging (LTA) of unmodified and modified bitumens were simulated using the rolling thin film oven (RTFO) and pressure aging vessel (PAV) tests respectively. The pull-off test was conducted using the universal testing machine (UTM). Stainless steel and granite substrates moulds were designed and fabricated for this purpose. Dry and wet conditions were simulated to evaluate the loss of bonding strength due to aging. Additionally, consistency tests, namely penetration test, softening point test, and viscosity test, were conducted on all binders. The activation energy (AE) for unaged and aged binders were also evaluated. The results for the consistency test; the pull-off test for stainless steel substrate; and the values for AE showed that the addition of the HNT decreased the effect of aging, which means that modified binders showed decreased temperature susceptibility and this is reflected in improved aging resistance. The result of the pull-off test on granite substrate showed that binders modified with HNT performed better under dry and wet conditions. It can be concluded that HNT is an anti-aging as well as anti-striping agent.

1. Introduction

Flexible pavements are affected by prevailing environmental factors such as sunlight, temperature fluctuation, moisture, oxygen, air voids, etc. These factors have profound effects on the mechanical properties of pavement mixtures, and they are the cause of most of the problems seen with pavements [1]. The aging of bitumen is one of the main factors affecting the properties of pavement mixtures through several types of mechanisms. One of the common mechanism that occurs in bitumen is oxidation [2]. Oxidation occurs when diffused oxygen reacts with bitumen. The rate of oxidation for the most part is determined by increase in temperature; the type of bitumen and its thickness; and the type of aggregate and its gradation [3]. Temperature increases oxidation rate during the mixing and compaction stages, and this is related to short-term aging. On the other hand, long-term aging occurs during the service life when the oxygen diffused in air voids reacts

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with the coated binder at the interface of the aggregate, causing it to oxidize. Oxidation increases viscosity, which in turn increase stiffness, and as stiffness increase, cracking begins to appear [4,5].

The fluid properties of a bituminous binder is determined by its components, which is related to the binder's chemical composition and the physical arrangement of the structure [6,7]. Bitumen is made up of four components, namely asphaltenes, resins, aromatics, and saturates. These components play an important role in determining the viscosity of binders. They are sensitive to aging that are caused by temperature increase. The increase in temperature changes bitumen's physical and chemical composition from semi-solid to fluid. The oxidation that occurs when bitumen is exposed to high temperature during the production process alters the bitumen's chemical composition. The increase in asphaltenes content causes a decrease in the resin and aromatic contents. These changes are reflected in the bitumen's physical properties and lead to what is called short-term aging [6]. In short-term aging, the changes in physical properties occur gradually due to the effect of high temperature (mixing and compaction temperatures), while in long-term aging the changes only occur after a long period of time (service life). The increase in asphaltene content causes bitumen to harden due to aging [8]. Temperature increase also increases the formation of the carbonyl and carboxylic groups, and these two groups also increase the hardness of bitumen. However, there are ways, other than aging, to increase binder's hardness; one way of doing this is by modifying binders through the addition of additives [9]. Increasing hardness through the modification of bitumen has the advantage of increasing stiffness, which in turn lead to better resistant towards thermal cracking, rutting, and fatigue [6,9–11].

Efforts are still being made to improve the aging resistance of bitumen [12,13]. Most studies which investigate the performance of bitumen and asphalt mixtures also investigate the effect of various stages of aging. Primary focus is given to improving the properties of unaged and aged binders and asphalt mixtures. Recently, focus has been shifted towards the use of nano-materials. Many studies have confirmed the benefits of using nano-materials as an additive to binders. For instance, Yang and Tighe [14] listed many benefits of using nano-materials as additives. Several types of improvements have been observed when nano-materials are used as additives. Adding nano-materials to bitumen increase the mixture's resistance to UV [15,16]. For example, adding nanoclay and micro-carbon fiber to bitumen significantly reduce the moisture susceptibility of the mixture [17]. The addition of nanoclay also significantly improved binder's adhesion force (which was evaluated using the atomic force microscopy (AFM) technique, with focus on the improvement in the nano- and micro-structures of the modified binder) [18]. The surface free energy (SFE) technique was used by Hossain et al. [19] to evaluate the effect of adding nanoclay to polymer modified bitumen. The result of this study showed an improvement in moisture susceptibility. Another advantage of using nanoclay is the expected cost saving of between 22% and 33% per metric ton of bituminous binder in comparison to using binders modified with polymer [19]. However the change in the internal structure of nanomaterial modified bitumen has not been fully understood and further investigation on the bonding strength of the mixture must be done using different kinds of techniques.

The bonding strength of bituminous binder is usually evaluated using the pull-off test method [20,21]. In 1963, Majidzadeh used a tensile machine to assess the tensile properties of bitumen [22]. Bonding strength was found to be sensitive to several factors, such as bitumen viscosity, aggregate type, curing temperature, curing time, and curing condition [21,23]. The pull-off test has also been effectively used to evaluate the changes caused by any kind of modification of the bitumen, such as the addition of additives, or in assessing the effects of aging [23,24]. Therefore, this study attempts to investigate the ability of pull-off test to evaluate the effects of aging on bitumen properties and to highlight the benefits of adding halloysite nanotube (HNT) to bitumen at various aging stages (unaged, short-term aging, and long-term aging). The pull-off test was used to determine the adhesion strength of unaged and aged binders. The consistency tests, namely the penetration, softening point, and viscosity tests, were used to evaluate the changes in binders before and after aging. The rotational viscosity test was conducted to evaluate the effects of HNT on the viscosity of the original binder and the aged samples. The activation energy (AE) was computed for samples under the same aging condition to further investigate aging effect. An investigation was also conducted to explore any correlation between the results for pull-off test, viscosity, and AE of the unaged and aged samples. Any correlations that exist indicate the benefits of using the pull-off test to evaluate the effect of aging on bituminous binders.

2. Experimental design

2.1. Materials and sample preparation

In this study, a 60/70 penetration grade bitumen was used as a control sample. The physical properties of the control binder were determined through the penetration, softening point, and viscosity tests. The details of its physical properties are given in Table 1. The halloysite nanotube (HNT) is a type of two-layered nanoclay, namely an alumina-silicate layer. Its chemical formula is $Al_4Si_4O_{10}(OH)_8{\cdot}4H_2O$. HNT particle was used to fabricate a tube which is 30–70 nm in diameter and 300–600 nm long (based on the datasheet of manufacture). The material was used in powder form and its colour ranges from white to tan. The field emission scanning electron microscopy (FESEM) was used to investigate the HNT particles. Fig. 1 shows the dimensional length of the HNT particles.

Table 1
Physical properties of base 60/70 penetration grade bitumen.

Test	Standard test	Value
Penetration (0.1 mm) at 25 °C	ASTM D5	66
Softening point (°C)	ASTM D36	46
Ductility (mm) at 25 °C	ASTM D113	100
Specific gravity at 25 °C	ASTM D70	1.03
Flash point (°C)	ASTM D92	310
Viscosity (mPa s) at 135 °C	ASTM D4402	551

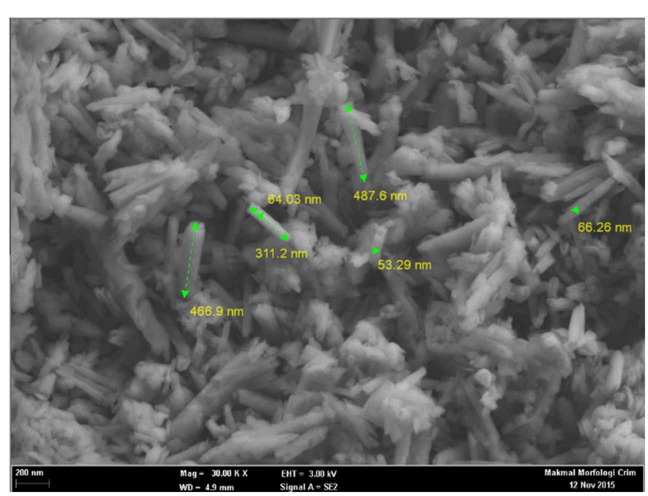

Fig. 1. FESEM of HNT particles.