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## Effect of silane coupling agent on improving the adhesive properties between asphalt binder and aggregates

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### HIGHLIGHTS

- SCA improves the adhesion between asphalt binders and aggregates.
- The adhesion and stripping energy were evaluated by contact angle tests.
- The tensile tests reveal the increased adhesion caused by SCA modification.
- The improvement of the adhesion is due to the formation of hydrogen bonds.

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### ABSTRACT

Moisture damage negatively affects the pavement service life. A typical way to delay the moisture damage is to improve the adhesion between asphalt binder and aggregates. The purpose of this paper is to investigate the effects of silane coupling agent (SCA) on improving the adhesion between asphalt binder and aggregates. In this paper, the contact angles of the aggregates modified by different SCA hydrolysates were measured. The adhesion energy and stripping energy between asphalt binder and aggregates modified by different SCA hydrolysates were calculated. Secondly, the tensile strengths between asphalt binder and aggregates before and after immersion in water were measured. Lastly, the interfacial interactions between SCA and the aggregates were studied by Fourier transform infrared (FTIR) and scanning electronic microscopy (SEM). The surface energy results prove that the SCA can significantly improve the adhesion energy and stripping energy between the aggregates and the asphalt binder. The tensile test results show that the tensile strength of the aggregates modified with a type of SCA (KH550) hydrolysate before immersion in water was increased by 58%, and the tensile strength after the immersion in water was increased by 155%. Based upon these results, it can be concluded that using SCA has a superior effect on improving adhesive properties between asphalt binder and aggregates.

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### 1. Introduction

Among the premature pavement failures, moisture damage is the most common and complex issue. When the water infiltrates into the asphalt-aggregate interface repeatedly, the asphalt binder gradually peels from the aggregate. This results in aggregate-shattering, pot holes and deformation. These pavement distresses

seriously affect the driving safety and comfort. Fundamentally, it is a priority to improve the adhesion between asphalt binder and aggregate to resist moisture damage [1–8].

Many approaches have been developed to improve the adhesion, including selection of aggregates [9], improvement of mixing techniques [10] and reduction of the impure dust-powder coatings on surface of the aggregates [11]. However, it is difficult to improve the adhesion between asphalt binder and aggregates using these methods. Recently, the incorporation of anti-stripping agents into asphalt mixture has been shown to be an effective method to resist moisture damage of asphalt mixture. In general, anti-stripping

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agents can be divided into four categories [12,13]. The first type is inorganic anti-stripping agent, mainly including lime and cement [14]. Such anti-stripping agents have the advantages of wide availability and low cost. Nevertheless, they are difficult to mix evenly in the asphalt mixture because of their powder shapes. In such case, the anti-stripping effect hardly works. The second type is the metallic soaps [15]. However, the density of these additives is quite different from that of asphalt binder, leading to the poor compatibility between additives and asphalt binder. The third type is surfactant, which is mainly quaternary ammonium salt [16]. This additive has a poor thermal stability, which tends to lose effectiveness at a high temperature. The fourth type is the high-molecular anti-stripping agent, which mainly includes amines and non-amine polymers [17]. The most common type is silane coupling agent (SCA), which has a better thermal stability than surfactant. SCA has a special molecular feature in which it has two different types of functional groups, including organofunctional groups and hydrolysable groups. SCA can easily bond silicate material and organic material due to this molecular feature [18]. Therefore, SCA has attracted a considerable amount of attention from many researchers. Ameri et al. [19] and Arabani et al. [20] studied the effect of a new nano-organosilane anti-stripping agent, called Zycosoi, on the asphalt-aggregate interface. They found that this anti-stripping agent effectively improves the adhesion between aggregate and asphalt binder as well as moisture damage resistance. Min et al. [21] developed a new anti-stripping agent based on KH570 and investigated its effect on the adhesion between aggregate and asphalt binder. The results showed that the new anti-stripping agent had distinctly improved the interface adhesion between asphalt binder and aggregate. Guo et al. [22] studied the effect of SCA on asphalt mixture and revealed that the SCA significantly improves the moisture resistance of the acid aggregate. Zhang et al. [23] found that the combination of titanate coupling agent and SCA significantly improves the adhesion between asphalt binder and aggregate. Liu et al. [24] found that the aluminate coupling agent enormously improves the anti-cracking property of asphalt mixture at low temperatures and anti-fatigue property at room temperatures, but it reduces the anti-rutting property of the mixture at high temperatures. Xie et al. [25] mixed SCA ( $\gamma$ -methacryloxypropyltriethoxysilane) and fly ash in asphalt mixture and found that this method affects the moisture damage resistance positively. Guo et al. [26] prepared a kind of nano-silica modified by SCA. The results showed that nano-silica improves the rutting resistance at high temperatures and anti-cracking performance at low temperatures. Wang et al. [27] disclosed that SCA forms Si—O—Si chemical bond between asphalt binder and aggregate and elevates Marshall stability and compressive modulus of the asphalt mixture by 10–30%. Liang and Guo [28] studied the effect of SCA (DB-570) on asphalt binders. They found that the asphalt binders on the aggregate surface modified by SCA is more uniform than that on the neat aggregate, indicating that the adhesion between asphalt binder and aggregate is enhanced. Mohd Hasan et al. [29] studied the effects of calcium carbonate and linear low-density polyethylene on the asphalt mixture. The results showed that the additive treated with the coupling agent only enhanced the resistance of moisture damage but not the anti-fatigue performance. Estevez [30] found that lecithin can improve the adhesion between rubber asphalt binder and aggregate. At present, these studies above have focused on the improving the adhesion property between asphalt binder and aggregate due to the usage of SCA. However, most of mentioned literatures only evaluate the adhesion performance by using mechanical experiments, which still lack a further explanation in terms of interfacial effects and microscopic theory.

The traditional evaluation methods of adhesion between asphalt binder and aggregate include boiling test, photoelectric

colorimetric test, freeze-thaw split test, et al. The surface energy test is a newly developed method. Cheng et al. [31] used the gas adsorption method and Wilhelmy plate method to measure the surface free energy between aggregate and asphalt binder respectively. They found that the surface free energy between asphalt binder and aggregate calculated by this method can help to select a more suitable asphalt-aggregate combination. Wei [32] measured the surface energy of aggregate and asphalt binder by using the sessile drop method and found that the calculated adhesion energy and the macroscopic tensile test results had a good consistency. Tan et al. [33] used the surface energy method to calculate the adhesion energy of different aggregates and different asphalt binders and pointed out that the modified asphalt binder had higher surface energy than raw asphalt binder. Arabani and Hamedi [34] studied the effect of liquid anti-stripping agent on asphalt binder and aggregate with a surface energy experiment and a dynamic modulus test. The results show that adding liquid anti-stripping additives causes the increase of total surface free energy of the asphalt binder and results in a decrease of stripping between the aggregate and asphalt binder in the presence of water. Similar results were obtained from a dynamic modulus test. Sung-Hee Kim et al. [35] revealed that moisture damage is caused by invasion of the moisture film into the asphalt-aggregate interface and the development of cracks under repeated loads. Kakar [36] studied the performance of chemical active agent with the surface energy method and found that anti-stripping agents reduce the surface energy of the aggregate and improve the moisture damage resistance of asphalt mixture. In addition, the moisture damage resistance of basalt is worse than that of limestone. Hamedi et al. [37] calculated interfacial adhesion energy by the surface energy method. They found that interfacial adhesion energy has a good correlation with macro-mechanical experimental results. Yan and Liang [38] conducted a self-developed shearing test to measure the adhesion between the Karamay K-90/Victory S-70 asphalt binder and granite/limestone, respectively. They found that the shearing test results and the boiled test results have a good consistency. Copeland [39] studied the adhesion performance between asphalt binder and aggregate by a modified pull-off test method. The destruction force was measured and the experimental results showed that the test method is useful. Zhang et al. [40] conducted the Vialit plate shock method to evaluate the adhesion of SBS modified bio-asphalt binders and aggregates at low temperature. They found that the SBS modified bio-asphalt mixtures had a higher low-temperature adhesion than the petroleum No. 50 asphalt binder mixture. Ji et al. [41] studied the influence of organic additives and water on the adhesion of the asphalt-aggregate interface according to the surface free energy theory. The results indicated that the addition of organic additives enhance the adhesion between the asphalt binder and the aggregate because organic additives reduce the surface free energy of the asphalt binder. Diab et al. [42] investigated the effects of nanosized hydrated lime and regular-sized hydrated lime on the asphalt binder rheology and evaluated their effects on the free energy of adhesion between the asphalt binder and the aggregate. They found that the measured free energies of adhesion are in agreement with the rheology results. From the researches above, it can be seen that the surface energy method can quantitatively evaluate the adhesion between asphalt binder and aggregates. However, there is still a lack of studies combining the surface energy analysis and tensile test to evaluate the adhesion properties between the asphalt binders and aggregates modified by SCA.

In this paper, the surface energy method is used to predict the adhesion between aggregates modified by SCA and asphalt binder. Secondly, the adhesion between asphalt binder and aggregates is verified by a direct tensile test and an immersion tensile test. Finally, the surface microstructure is used to explain the essence

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