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## The anti-icing and mechanical properties of a superhydrophobic coating on asphalt pavement



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#### **HIGHLIGHTS** highlights are the second control of the secon

A superhydrophobic coating delays icing time of water droplets.

Adhesion force between ice and a superhydrophobic coating decreases.

Fumed silica improves the tensile strength of RTV.

KH550 increases adhesion force between SC and asphalt mixture.

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### **ABSTRACT**

Ice formation on asphalt pavement may lead to potential hazards for driving safety. To reduce the safety risk, this paper aims at preparing a superhydrophobic coating (SC) on asphalt pavement. Roomtemperature vulcanized silicone rubber (RTV) is the main component of the SC. The micro-/nano- $SiO<sub>2</sub>$ particles modified by a silane coupling agent (KH550) were sprayed on the RTV surfaces to prepare the SC. The contact angle test and surface morphology observation results indicate that the contact angle of water droplets increases effectively due to the rough micron-/nano- structures on the SC. Additionally, icing observation reveals that SC delayed icing time of water droplets 1.5 times longer than the asphalt mixture specimen without SC. Adhesion force between ice and SC decreases by 84% compared with that between the ice and asphalt mixture at  $-5$  °C. The tensile strength of the RTV sheet containing 30 parts per hundred parts (phr) of fumed silica is 13 times than that of the RTV sheet without fumed silica. Adhesion force between SC and asphalt mixture achieve the maximum value of 875 kN when SC contains 5 phr of KH550. To sum up, it can be concluded that the SC has superior anti-icing and mechanical properties on asphalt pavement.

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#### 1. Introduction

In view of the icing problems on asphalt pavements, the frequently-used methods of removing ice currently include spraying chloride ice-melting agents, manual removal, mechanical removal and solar-energy utilization. Nevertheless, the methods above will either aggravate pollutants, corrode metal structures, reduce the performance of asphalt binders or be quite expensive  $[1–11]$ . The reason why ice is difficult to completely remove from asphalt pavement is because ice will be embedded in the gaps inside asphalt mixture to form an anchor structure after the water has penetrated into the voids of the asphalt pavement and frozen. This anchor structure greatly enhances the adhesion of ice on the asphalt pavement  $[12-15]$ . Moreover, surface energy of the pavement also affects adhesion between the ice layer and the asphalt pavement, according to Quasi-liquid layer theory [\[16–18\].](#page--1-0)

Enlightened by the self-cleaning ability of superhydrophobic structures on plant surfaces such as lotus leaf and rice leaf [\[19,20\]](#page--1-0), researchers have recently focused on constructing SC to

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delay icing process on material surfaces in cold environments. Generally, a certain material is defined as a superhydrophoc material when the contact angle ɵ of a drop on its surface surpasses 150. Because of a smaller contact area between drop and superhydrophobic material [\[21,22\]](#page--1-0), as well as the delayed speed of ice formation from a water droplet and the inferior adhesion of the ice layer, research on the utilization of SC to improve surface ice formation have aroused general interests in recent years.

Saito et al. [\[23\]](#page--1-0) manufactured a superhydrophobic waterproof coating by mixing micron-sized tetrafluoroethylene with polyvinylidene difluoride. Through ice adhesion experiments, it was found that the adhesion of the ice layer increased with the roughness of SC. Nakajima et al. [\[24\]](#page--1-0) studied the basic theory of surface wettability of SC and described the progress in the processing and performance of the SC. Wang et al. [\[25\]](#page--1-0) prepared four infiltrating aluminum surfaces from superhydrophilic to superhydrophobic by a combination of etching and coating methods. The results showed that the superhydrophobic aluminum surface has a larger contact angle but a smaller rolling angle than superhydrophilic aluminum surface, and the adhesion of ice layer on superhydrophobic aluminum surface is only 13% of that on superhydrophilic aluminum surface. Arianpour et al. [\[26\]](#page--1-0) developed a SC with a contact angle greater than  $150^{\circ}$  by spraying a diluted hexane suspension doped with nano-powder on an aluminum substrate surface. The experimental results showed that the SC significantly delayed the freezing process of surface water droplets. Pan et al.  $[27]$  prepared a SC with a contact angle of 158 $\degree$  and excellent ice protection as well as corrosion resistance by spraying PMMA and hydrophobic silica on the metal surface. Farhadi et al. [\[28\]](#page--1-0) studied the anti-icing performance of micro-/nano-structure SCs with different chemical surfaces and morphologies by testing the adhesion strength of ice layer on the surface of SCs. The experimental results showed that the icing/de-icing frequencies and ambient humidity both affect the adhesion strength of the ice layer. Mishchenko et al. [\[29\]](#page--1-0) constructed a micro-/nanostructured superhydrophobic material and studied the icing conditions of dynamic water on its surface. The results showed that the surface of the superhydrophobic material can be designed as ice-free because the dynamic water repels before ice could crystallize. Kulinich et al. [\[30,31\]](#page--1-0) tested the adhesion of ice layers on the hydrophobic/superhydrophobic coating surfaces with similar chemical compositions by simulating outdoor icing conditions. They found that the adhesion of the ice layer was related to the wetting hysteresis of the water droplets on the SC surface. Cao et al. [\[32\]](#page--1-0) studied the anti-icing performance of a SC made of a nanoparticle polymer composite and found that the anti-icing ability of the composite depended not only on its superhydrophobicity, but also on the sizes of the particles exposed on the coating surface. The critical particle sizes, which determine the superhydrophobicity and anti-icing properties, were divided into two distinct length scales. The studies above have shown that SC has desirable superhydrophobic properties and they can have an adequate anti-icing effect when it is on the surface of materials. However, few relevant research focuses on the influence regarding anti-icing performance of SC on asphalt pavements.

Since asphalt pavement is affected by vehicle loads, anti-icing coatings on asphalt pavements are expected to satisfy some mechanical properties. Because of friction of tires applied to the asphalt pavement, the mechanical properties of the anti-icing coating include the tensile strength of coating and the adhesion strength between coating and asphalt mixture. However, the backbone of silicone rubber is quite soft and the interaction between molecular chains is weak, the strength of the raw RTV is required to be reinforced by adding some fillers. Fumed silica is the most effective reinforcing filler to enhance the tensile strength of RTV coatings [\[33\].](#page--1-0) Donnet [\[34\]](#page--1-0) found that fumed silica particles whose size is greater than 1 um do not substantially reinforce the silicone rubber and increase the viscosity of the silicone rubber. In contrast, fumed silica particles whose size is less than 100 nm have a sufficient reinforcing effect on the silicone rubber. Wang [\[35\]](#page--1-0) demonstrated that the network structure formed between particles increases the effective volume of the filler reinforcement, which can further improve the modulus of the elastomer. Meanwhile, since the surface of silica contains a large amount of hydroxyls, it exhibits a strong hydrophilicity and results in forceless bond with the silicone rubber. Therefore, silica surface needs to be modified to improve its compatibility and fluidity in silicone rubber. Wang [\[36\]](#page--1-0) studied the mechanical and adhesion properties of silicone rubber with addition of silane coupling agents (SCA) including A-172, A-151 and A-189. The results showed that both A-172 and A-151 can strengthen the mechanical and adhesion properties of silicone rubber and the optimal dosage of SCA is between 2phr and 4phr. The above studies have investigated the mechanical properties of SCs under various circumstances. However, the mechanical properties of SCs subjected to vehicle loads on asphalt pavement still need to be further tested.

From Young's equation and Wenzel's theory [\[37\]](#page--1-0), it can be concluded that preparation of materials with higher contact angles largely depends on constructing material surfaces with appropriate roughness densities to reduce the surface energies. A specific method is to construct a micro-/nano- structure on material surfaces with low surface energies.

In this paper, a SC was prepared by spraying micron/nano- $SiO<sub>2</sub>$ particles on RTV surfaces. Subsequently, anti-icing tests of SC on asphalt pavement were conducted by freeze observation tests on asphalt mixture and tensile tests between ice layer and asphalt mixture. Eventually, tensile strengths of RTV sheets and adhesion forces between SC and asphalt mixtures were measured to evaluate the mechanical properties of SC. The flowchart of this research is shown in [Fig. 1.](#page--1-0)

#### 2. Experiments

### 2.1. Raw materials

Silicone rubber is the main component of uncured roomtemperature vulcanized silicone rubber. The catalyst used is industrial-grade dibutyltin dilaurate. The particle size of microsilica particle is 1  $\mu$ m $\sim$ 5 $\mu$ m. A coupling agent is KH550 [(3-Amino propyl)triethoxysilane] with a purity of 97%. The density (25  $\degree$ C) of methylsilicone oil is 0.955 kg/m<sup>3</sup>-0.975 kg/m<sup>3</sup>. The particle size of fumed silica is 20 nm and its specific surface area is 200  $m^2/g$ . The physical properties of asphalt binder are shown in [Table 1](#page--1-0). Aggregate used is basalt.

#### 2.2. Modification of  $SiO<sub>2</sub>$  particles and FTIR test

KH550, ethanol, and water were blended at a mass ratio of 1:10:1. The pH value of the solution has to be adjusted between the range of 8 and 10 due to amino functional groups. Then the solution was transferred to an oven to be hydrolyzed at 40  $\degree$ C for a half hour. After hydrolysis, the micro-/nano- $SiO<sub>2</sub>$  particles were added to the solution and well mixed. The temperature of the oven was kept at 140 °C for 2 h to obtain the modified micro-/nano-SiO<sub>2</sub> particles. In this process, hydroxyl groups on the surface of the  $SiO<sub>2</sub>$ particles reacted with silanol formed by the hydrolysis of KH550, condensing to form stable -Si-O- covalent bonds. The reaction process is shown in [Fig. 2.](#page--1-0)

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