Construction and Building Materials 171 (2018) 214–222

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Impact of minerals and water on bitumen-mineral adhesion and debonding behaviours using molecular dynamics simulations

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HIGHLIGHTS

- Bitumen-mineral adhesion decreases from microcline, albite, calcite to quartz.
- Moisture reduces more adhesion for neutral minerals than alkali minerals.
- van der Waals dominates bitumen-mineral adhesion for neutral minerals (quartz)
- Electrostatic interaction dominates bitumen-mineral adhesion for alkali minerals.
- Bitumen-mineral debonding is a thermodynamically favourable process.

ARTICLE INFO

Article history:
Received 24 January 2018
Received in revised form 12 March 2018
Accepted 19 March 2018

Keywords:
Aggregate minerals
Bitumen-mineral interface
Adhesion
Debonding
Stripping mechanism
Molecular dynamics

ABSTRACT

This study aims to evaluate the effects of mineral types and water on the adhesion properties and debonding behaviours of bitumen-mineral interface systems. A molecular dynamics modelling approach was employed to simulate the interactions between minerals and bitumen with and without the presence of water. Four representative minerals (quartz, calcite, albite and microcline) were selected to build the mineral-bitumen interface systems and the mineral-water-bitumen interface systems in the molecular dynamics models. The adhesion property between minerals and bitumen was quantified by work of adhesion, defined as the energy required to separate a unit area of the bitumen-mineral interface. The debonding behaviour between minerals and bitumen is characterised by work of debonding, defined as the energy required to displace bitumen by water at the mineral-bitumen interface. The simulation results were validated by available experimental results reported in the literature. It was found that the work of adhesion and the work of debonding for the four bitumen-minerals interface systems are ranked microcline > albite > calcite > quartz at both dry and wet conditions. Moisture can reduce the adhesion between minerals and bitumen by 82%, 84%, 18% and 1% for the quartz, calcite, albite and microcline, respectively. The adhesion between minerals and bitumen is attributed to the non-bond interaction energy, in which the major component is van der Waals interaction for neutral minerals (e.g., quartz) and the electrostatic interaction for the alkali minerals (e.g., calcite, albite and microcline). The bitumen-mineral debonding is a thermodynamically favourable process with reduced total potential energy of the system. It is concluded that the bitumen-mineral adhesion and debonding behaviours strongly depends on the chemistry and mineralogical properties of the minerals. This work provides a fundamental understanding of the adhesion and debonding behaviours of the bitumen-mineral interface at the atomistic scale.

1. Introduction

Asphalt pavements are subjected to heavy and repeated traffic loading, leading to severe cracks and moisture damages when water exist in the pavement structures. Zhang and Lytton et al.

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https://doi.org/10.1016/j.conbuildmat.2018.03.136
0950-0618/

investigated the crack initiation and evolution in asphalt mixtures under compressive and tensile loads [1,2]. They found that the bond energy of an asphalt mixture dominated the crack initiation and propagation processes and it increases with aging period and loading rate and decreases with temperature [3,4]. Previous studies have reported that the mechanical properties of asphalt mixtures had a strong dependency upon the interfacial bond between bitumen and aggregate [5,6]. Therefore, the adhesion of the bitumen-aggregate interface needs to be strong and durable under the complex traffic and environment conditions. Fundamentally this is caused by that water diffuses into asphalt layer of the pavements and weakens the bitumen-aggregate adhesive bond in asphalt mixtures. Stripping happens when the adhesive debonding occurs between bitumen and aggregate. In order to understand the stripping mechanism of asphalt mixtures and extend the pavement service life, there is a fundamental need to investigate the adhesion properties and moisture damage mechanisms of the bitumen-aggregate interface.

Researchers have developed experimental testing methods to investigate the mechanism of adhesion between bitumen and aggregate with and without moisture effect. Lytton et al. [7] measured surface energy components of bitumen and aggregate and derived the adhesive bond strength between aggregate and bitumen to quantify the moisture susceptibility of the asphalt mixture. Khattak et al. [8] performed the lap-shear test to study binder-aggregate adhesion at low temperature. Canestrari et al. [9] used the Pneumatic Adhesion Tensile Testing Instrument (PATTI) to evaluate the influence of water on cohesive and adhesive properties of bitumen-aggregate systems. Moraes et al. [10] investigated the feasibility of the Bitumen Bond Strength (BBS) test for moisture damage characterization of the bitumen-aggregate interface. Fini and Al-Qadi [11] developed a pressurized blister test to characterize the interfacial adhesion between aggregate and bituminous materials. Fischer et al. [12] employed atomic force microscopy (AFM) to investigate the interfacial interaction between bitumen and the minerals as present in asphalt mixtures. These experimental studies provided a good understanding of bitumen-aggregate adhesion and moisture susceptibility of the asphalt mixtures.

Recently, with the development of the high-performance computation, atomistic and molecular modelling has become an effective way to interpret the material properties or system behaviours from fundamental molecular processes. Researchers employed molecular dynamic (MD) simulation to investigate the molecular interaction and basic properties of bitumen and the mechanical behaviours of asphalt concrete. Zhang and Greenfield [13,14] and Li and Greenfield [15,16] developed the model bitumen systems to represent the real bitumen and analysed the physical properties of the model bitumen (e.g. viscosity, relaxation time, diffusion coefficient, and dynamics) using molecular simulation. Lu and Wang [17,18], Wang et al. [19], and Xu and Wang [20] performed MD simulation to evaluate the mechanical properties of the bitumen-aggregate interface. MD simulation has also been applied to investigate the oxidative aging behaviours [21–23], diffusion and self-healing mechanisms [24–26], and micromechanical properties of bitumen [27–29]. These research efforts have produced important results for better understanding the physical, rheological, and thermodynamic properties of the real bitumen from a more fundamental perspective.

However, most of the research effort was spent on the effect of the bitumen or bitumen compositions on the adhesion, whereas little work has been focused on the effect of aggregate mineral compositions on the adhesion and debonding behaviours between aggregate and bitumen at the atomistic scale. Some experimental studies have shown that the chemical and mineralogical compositions of the aggregate minerals significantly affect the stripping properties of asphalt mixtures. Lyne et al. [29] reported that the elemental composition of a mineral affected its dispersive adhesion to bitumen. Horgnies et al. [30] presented that the adhesion of the bitumen on micas and quartz was better than the one on alkali feldspars. More fundamental studies by atomistic modelling are needed to reveal the mechanisms of the debonding between aggregate minerals and bitumen when different mineral compositions are present.

The objective of this study is to investigate the effects of aggregate mineral types on the adhesion property and debonding behaviour of the bitumen-mineral interface with and without the presence of water using an atomistic modelling approach. The molecular models of four representative minerals were employed to construct the mineral-bitumen and mineral-water-bitumen interface systems. The interface molecular interactions were quantified using the molecular dynamics (MD) simulations. The adhesion and debonding properties of the bitumen-mineral interface were evaluated using the work of adhesion and the work of debonding. Furthermore, the wet interface stripping mechanism was analysed through investigating the mineral-bitumen debonding process at the atomistic scale.

2. Aggregate minerals

Aggregates used in pavement structure are mineral rocks. The composition of the aggregates, based on chemistry and mineralogy, are quite complex and have diverse mineralogical composition. Some aggregates mainly contain one mineral such as quartzite (SiO_2) and limestone ($CaCO_3$). Other aggregate types are composed of several different minerals. For example, the major minerals in granite are quartz, mica and feldspar.

A mineral is an element or chemical compound that is normally crystalline and has been formed as a result of geological processes [31]. The eight most common elements of the minerals in the earth's crust are oxygen, silicon, aluminium, iron, calcium, sodium, potassium and magnesium [32]. Natural rock mineral is polycrystal with local periodicity. A crystal structure is based on the orderly internal atomic or ionic arrangement. The structure of a mineral in the geometric form is often expressed by a box with parallel sides called the unit cell. The box with periodicity in all three dimensions is defined by six lattice parameters (lengths a, b, c, and angles α, β, γ). The crystal structure greatly affects a mineral's physical properties, and it together with the mineral chemical composition defines a mineral. Therefore, a mineral has a characteristic chemical composition and a regular atomic structure, leading to specific physical properties.

In this study, four representative minerals (quartz, calcite, albite, and microcline) were selected to investigate their debonding behaviour with bitumen using MD simulation. These four minerals were selected based on that 1) they are the common minerals existing in the aggregates that are used for constructing asphalt mixtures and 2) the four minerals have been comprehensively measured for their surface energy in the literature by Little and Bhasin [33].

3. Simulation models and methods

3.1. Molecular dynamics simulation

Molecular dynamics (MD) is a computational modelling technique for analysing the fundamental material properties such as density, modulus, viscosity, diffusion and surface energy and the material performance such as deformation and debonding at nanoscale through simulating the material atomistic movements and interactions. The principles of MD simulation are statistical mechanics and Newton's law of motions. MD is used to calculate