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Molecular dynamics study of interfacial mechanical behavior between asphalt binder and mineral aggregate

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HIGHLIGHTS

- Derive cohesive law based interface failure model using MD simulations.
- Evaluate effects of loading rate and temperature on interface failure behavior.
- Investigate chemo-mechanical relationship of interface failure.

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ABSTRACT

This study aims to study the deformation and failure behavior of the asphalt-aggregate interface using molecular dynamics (MD) simulations. The 12-component asphalt molecular models and the hydrated silica substrate were employed to form a bi-material interface system. Physical properties of asphalt binder were predicted from MD simulations including density and glass transition temperature for model validation with experimental data. Tensile simulations were performed and the stress-separation responses were obtained to analyze the interfacial mechanical behavior. It was found that the interface failure was mainly adhesive failure although large air voids were formed in the bulk asphalt as the loading rate decreases to a certain level. The interface failure strength and post-peak deformation are affected by loading rate and temperature that is consistent the viscoelastic behavior of asphalt binder. The stress-separation responses match the cohesive zone model (CZM) model that is usually observed in the pull-off strength test at the macroscopic scale. The relationship between chemical compositions of asphalt binder and the interface failure parameters was investigated. The MD simulation shows promising results to understand mechanical failure of asphalt-aggregate interface at the atomistic scale.

1. Introduction

Asphalt mixture is widely used in pavements of roadway, airport, and bridge deck. Asphalt mixture is a composite material consisting of mineral aggregate bound together with asphalt binder. Asphalt-aggregate adhesion is considered to be critical for the durability of asphalt pavements under traffic loading and environmental conditions. The bi-material interface creates the weak link in the composite asphalt mixture. Interfacial imperfection characteristics have significant effects on the overall response of composite material, such as mechanical properties and failure strengths of asphalt concrete [1]. Therefore, characterization of asphalt-aggregate interface properties is prevailing among many properties of asphalt mixture.

Experimental works have been carried out extensively to measure asphalt-aggregate interface failure behavior. Optical microscope (OM) image and scanning electron microscope (SEM) were adopted to probe the interfacial morphology [2]. X-ray computed tomography (CT) was used to study the 3-dimensional (3D) distribution of the asphalt mastic in fine aggregate mix and asphalt concrete [3]. Atomic force microscopy (AFM) was employed to investigate the interfacial interaction and adhesion strength between asphalt binders and mineral surfaces [4,5]. Dynamic Mechanics Analyzer (DMA) testing [6] and Dynamic shear rheometer (DSR) testing [7] were carried out to characterize the adhesion bond strength and adhesion fatigue performance of asphalt concrete composites under shear loading. Pull-off tests were performed by pneumatic adhesion tensile testing instrument (PATTI) and universal testing machine (UTM) to study the interface tension failure between asphalt and aggregate with and without moisture [8,9].

Although experimental testing methods provide effective approaches to measure adhesive strength between asphalt and

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aggregate at different scales, it has to be noted that carrying out experiments is known to be time and labor consuming. The measured interface behavior is affected by aggregate surface conditions in the specimen preparation process, such as texture, porosity, moisture, and contamination. On the other hand, asphalt is a complex material with many different types of chemical compounds and functional groups, while aggregate is normally crystalline with different mineral components resulted from geological processes. The adhesive interface behavior between asphalt and aggregate is strongly dependent on intermolecular interactions between different molecular groups [10]. Challenges exist to understand how chemical compositions of asphalt binder or mineral aggregate affect the mechanical behavior of asphalt-aggregate interface [11,12].

The emerging field of nano-mechanics is providing a new approach for study of the mechanics of composite materials, particularly simulating fundamental atomic mechanisms involved in the initiation and evolution of damage at material interfaces in the heterogeneous material. Molecular dynamics (MD) simulation is a powerful tool to describe material behaviors at atomistic level and characterize the relationship between chemical structure and engineering properties of material. Even though asphalt material is a complex chemical mixture and detailed information on its chemical components is hard to obtain, efforts have been made and important improvements have been achieved using MD simulations in recent years. Previous researchers have developed several atomistic models for asphalt binders and calculated thermodynamic properties including density, diffusivity, and viscosity based on MD simulations [13–15].

However, limited researches were found studying the interfacial mechanical behavior between asphalt and aggregate using MD simulations. In one relevant work, atomistic simulation was used to study asphalt-silica interface failure behavior; however, the adhesion strength could not be determined due to high stress variations in the interface region [16]. Although the atomistic-scale simulations can be potentially used to study the interface failure mechanisms such as dislocations, cracking, or sliding, it is critical to adopt accurate molecular models and utilize rational inter-molecular and intra-molecular potential field in the simulations. Therefore, more efforts are needed to develop systematic molecular modeling procedures for asphalt binder and mineral aggregate and study their interface failure behavior.

Therefore, the purposes of this study aims are to 1) investigate the deformation and failure behavior of asphalt-aggregate interface at the atomistic level using MD simulations and 2) discover the chemo-mechanical relationship of interface properties. The 12-component asphalt molecular models and the hydrated silica substrate were employed to form a bi-material interface system. Physical properties of asphalt binder were predicted from MD simulations and compared with experimental data in the literature for model validation. The velocity loading algorithm was applied for tensile simulation to obtain the stress-separation curve at the interface. Parametric studies were conducted to see the effects of loading rate, temperature, and unloading on the interface failure behavior. The chemo-mechanical linkage between the chemical compositions of asphalt binder and the interfacial adhesion strength and work of separation was investigated.

2. Molecular dynamics simulation method

2.1. Molecule models of asphalt

Asphalt is produced through petroleum refining process, and the elemental composition of asphalt depends primarily on its crude oil source. Asphalt has a high hydrocarbon content (>90% in weight), and various chemicals are found in asphalt, which results in the complexity of asphalt chemistry. Modern separation techniques divide asphalt into different fractions, which had similar polar and molecular characterization. Different separation methods cause different fractions of asphalt. From solubility point of view, asphalt materials are composed of three main constituents, i.e., asphaltene that is the most viscous and polar components; saturates, consisting of aliphatic molecules and are least viscous and non-polar; and resin whose properties are in between of above two. Besides, a four components concept was proposed by American Society for Testing and Materials (ASTM) D4124-09, in which asphalt is divided into saturate, aromatics, resins and asphaltenes four components. It can be observed that resins in the three components of asphalt were further divided into resins and aromatics in the four components of asphalt.

Recently, with the development of computer simulation and material characterization method, three 12-component asphalt models were proposed by Li and Greenfield [14] to represent three of SHRP core asphalt, namely are AAA-1, AAK-1 and AAM-1 asphalts, in order to better understand asphalt physical, rheological and mechanical properties. Larger molecules than three-component asphalt systems in the past can improve agreement with size characterization data, and by using molecular simulations, it is supposed to provide us a better understanding of asphalt physical, rheological and mechanical properties. For this study, these newly developed models were used as they are. Each separate molecule structure is illustrated in Fig. 1.

The numbers of each component in the asphalt model were chosen by the comparison between model and experimental values for all three asphalt systems, as shown in Table 1.

2.2. Molecule models of aggregate

Silica, or silicon dioxide, is used in this study to represent the aggregate or mineral, due to its simple chemical composition and availability of experimental data. In addition, silica makes up a large proportion of aggregate and usually more than 40% by weight, e.g., 72% of granite stone which is a widely used aggregate in pavement industry. Therefore, it is assumed that the structural behavior of asphalt-aggregate interface can be interpreted by studying the asphalt-silica interface in an atomistic scale.

A unit cell of silica has a lattice parameters of a = 4.913 Å, b = 4.913 Å, c = 5.405 Å, $\alpha = 90°$, $\beta = 90°$, $\gamma = 120°$, which is shown in Fig. 2(a). To model the silica substrate, the crystal is first cleaved in a way that the normal vector of the cleaved surface is at [0 0 1] direction, and the thickness of surface is larger than non-bond cut-off distances which is descried in following section. After relaxing the surface, the surface area was increased by repeating in x and y directions. After that, a vacuum slab was added to change the periodicity from 2-dimension to 3-dimension.

Furthermore, terminating hydroxyl (OH) groups with a density of 4.5 OH group/nm^2 were added to represent a fully hydrated silica film [19]. This is because surface silica is very reactive and will be hydroxylated in presence of water in practice [20]. Fig. 2(b) shows the final hydrated silica model to be used for simulation.

2.3. Force field

Interactions within and between asphalt and silica are formulated through the definition of force field. Optimized Potentials for Liquid Simulations (OPLS) all-atom force field, which is popular for simulating biomolecules and polymers, is adopted in this study. Its functional form is simple and parameters are typically determined by quantum chemical calculations combined with thermophysical and phase coexistence data.