

Analysis of foaming mechanism and adhesion of foam rubberised asphalt from the perspective of molecular scale

Kun Xiong¹, Jiupeng Zhang^{1*}

(1. School of Highway, Chang'an University, Xi'an 710064, China)

Abstract: Rubberized asphalt (RA) is characterized by its high viscosity, which necessitates elevated construction temperatures. During the mixing, paving, and compaction processes of the mixture, it consumes a significant amount of energy and releases gases harmful to both the environment and human health, thereby impeding the sustainable development of society. Foaming technology can effectively ameliorate this issue. However, RA exhibits a low foaming expansion rate, and the foaming temperature and water content have a considerable impact on the foaming efficacy. Current research on the foaming effect of RA is predominantly based on the macroscopic performance of test indicators, and the understanding of the microscopic foaming mechanism remains to be further elucidated. Additionally, some experiments have shown that foaming water can alter the microstructure of asphalt, adversely affecting the fatigue resistance and bonding strength of the asphalt binder. It is imperative to investigate the influence of water molecules on the interaction between foamed rubberized asphalt and aggregates. Therefore, it is essential to explore the foaming mechanism of rubberized asphalt by means of molecular dynamics and to evaluate the interaction characteristics between foamed rubberized asphalt and aggregates.

In this study, Foamed rubberized asphalt (FRA) models were constructed by incorporating water molecules and rubber chains into the asphalt model. The foaming process was comprehended by calculating and analyzing the density and energy changes of the model. The variation patterns of the foaming expansion rate and half-life of FRA were discussed through model parameters, and the Fourier transform infrared spectroscopy of RA before and after foaming was tested. The distribution of water molecules and the aggregation characteristics between molecules in the model were further explored through model conformation and radial distribution function. The mechanism of the influence of water molecules on the viscosity of the model was elucidated. Moreover, mineral models of granite and limestone were developed using SiO₂ and CaO supercells, respectively, and were combined with the FRA model to create interface models. The migration patterns of asphalt components and rubber molecules on the mineral surface were studied, and the effect of water molecules on interface adhesion was investigated through adhesion work.

The study reveals that an increase in temperature and the number of water molecules can reduce the density of the model. Furthermore, the time required for the model to reach a steady state is strongly influenced by the number of water molecules, indicating that water molecules interfere with the movement of other molecules within the model. Figure 1a elucidates the aforementioned conclusion from an energy perspective. It can be observed that an increase in temperature facilitates a rise in the total energy of the model, while an increase in the number of water molecules primarily contributes to the kinetic energy of the molecules within the model. The instability of the model makes volume compression difficult, which can explain the expansion mechanism of FRA. In other words, the more challenging the volume compression, the easier the expansion process, corresponding to an increase in the expansion rate. During the NPT process, the model compresses from a low-density steady state to a high-density steady state, which can reflect the decay process of FRA (Figure 1d). The energy difference between the two steady states is calculated in Figure 1b. It can be observed that the energy difference increases with water content at different temperatures. This suggests that the FRA model releases more energy when transitioning from a low-density state to a high-density state. This can explain the decay mechanism of FRA. In the actual foaming process, an increase

in foaming water produces more asphalt foam, leading to a shorter half-life. This is attributed to the thermodynamic instability of the foam system, which results in greater energy release. Changes in chemical bond energy and FTIR results further indicate that the foaming process does not significantly affect the chemical structure of FRA.

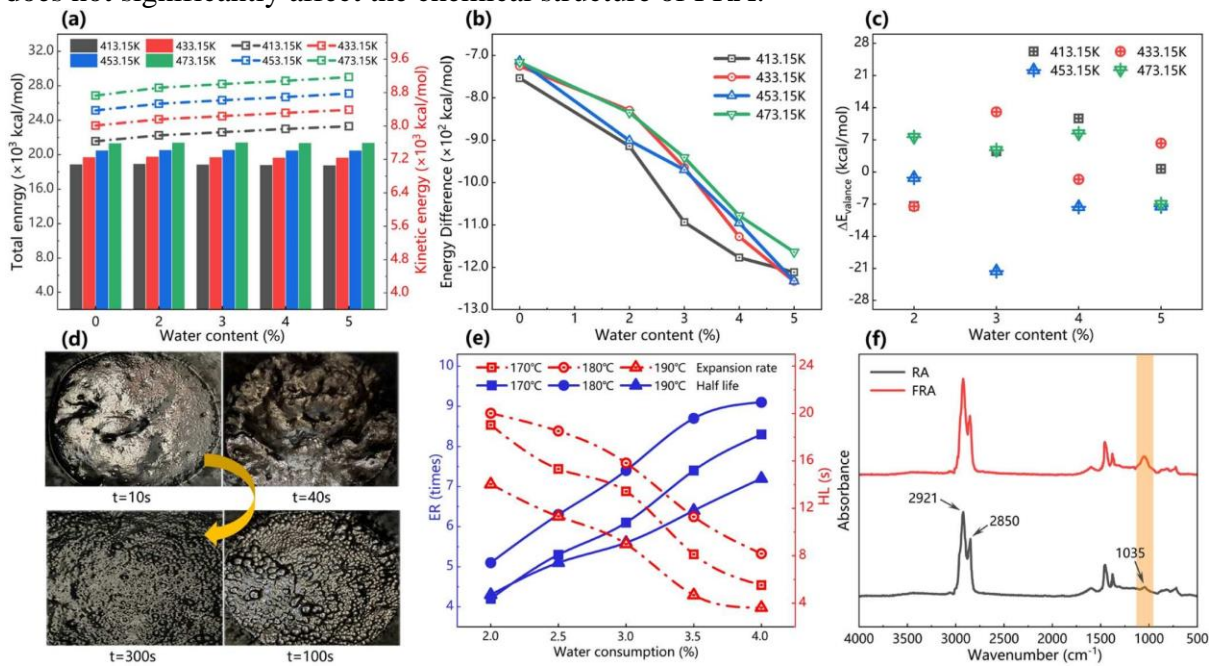


Figure 1 (a) Total energy and kinetic energy of the model; (b) Difference in energy at volume change; (c) Energy changes associated with chemical bonds in the model; (d) FRA decay process; (e) Experimentally obtained foaming expansion and half-life of RA; (f) FTIR images of RA before and after foaming.

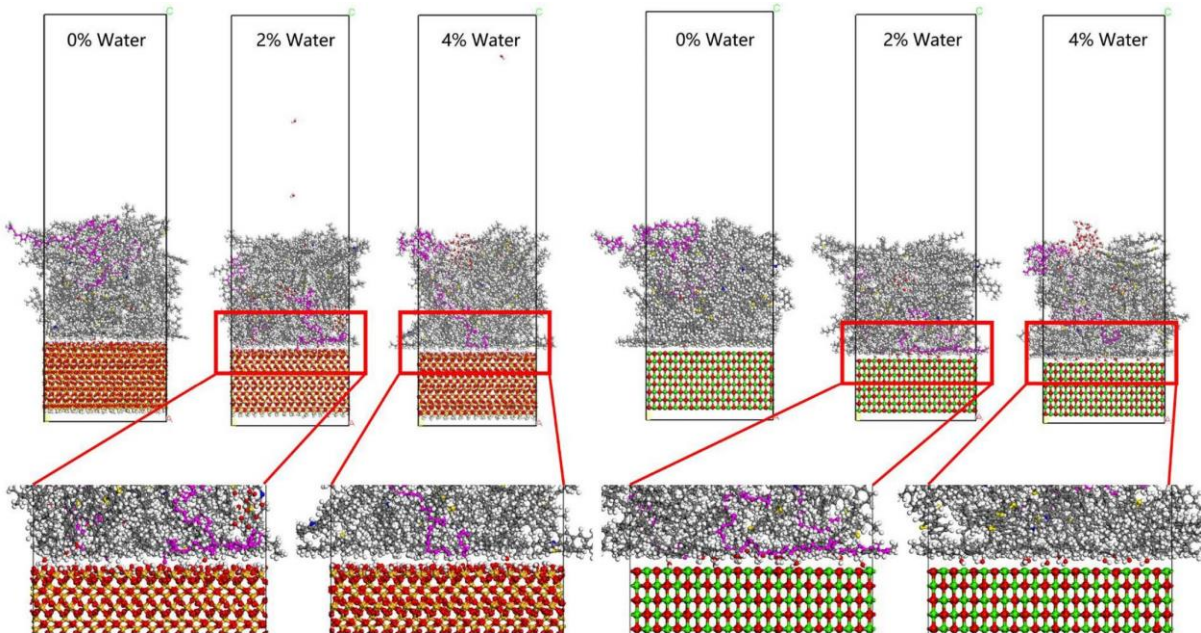


Figure 2 Interfacial conformations of FRA with minerals at different water contents.

In the interface model, it was found that there is no significant relationship between the number of water molecules and the interfacial adhesion work, which differs from the experimental results. This phenomenon can be well explained using molecular conformation. Figure 2 displays interface models with varying water content. It can be observed that water molecules exist in aggregated forms within the FRA and also in a diffused state at the interface. The water molecules inside the FRA may be related to its cohesive force, while the water

molecules at the interface can affect the adhesion between FRA and minerals. A study has shown that when water molecules intrude into the interface between RA and minerals, they weaken the interaction between the H atoms in RA and the O atoms in the minerals. This indicates that water molecules indeed disrupt the adhesion between FRA and minerals. However, the water molecules in FRA diffuse randomly. On one hand, the initial position of the water molecules determines the ease with which they can diffuse to the interface. On the other hand, water molecules tend to form hydrogen bonds with each other, aggregating and limiting their diffusion. Therefore, an FRA model with more water molecules does not necessarily have more water molecules at the interface. The conformation of the interface confirms this. Additionally, water molecules that diffuse to the interface find it difficult to form a water molecule layer that separates FRA from mineral molecules, which is also one of the reasons why the adhesion work is not applicable. This study provides a molecular interpretation of the foaming mechanism of RA and highlights the limitations of using adhesion work as an evaluation metric, offering valuable insights for guiding the foaming design of RA.

Keywords: Pavement engineering; rubberised asphalt; molecular dynamics; foaming mechanism; adhesion performance

Short Abstract

Analysis of foaming mechanism and adhesion of foam rubberised asphalt from the perspective of molecular scale

Kun Xiong ¹, Jiupeng Zhang ^{1*}

(1. School of Highway, Chang'an University, Xi'an 710064, China)

Abstract: Rubberised asphalt faces challenges such as viscosity and construction energy consumption. These problems can be ameliorated by the use of foaming technology. However, existing studies lack a microscopic-level understanding of the foaming mechanism and interfacial properties of foam rubberised asphalt. Therefore, this study used molecular dynamics simulation methods to establish foam rubberised asphalt models and interface models. The models were analysed from the aspects of model density, energy change, molecular agglomeration behaviour and adhesion work. Results revealed that increased temperature and water consumption caused difficulties in the volumetric compression of the model. The diffusion and aggregation behaviour of water molecules altered the distribution of other molecules in the model, effectively reducing the viscosity of the rubberised asphalt without affecting the chemical structure. In the interfacial model, water molecules contribute to the migration of asphalt molecules to the mineral surface, which exists in two forms: aggregating within asphalt and diffusing to the interface. The interface water molecules enhance the electrostatic interaction between asphalt and minerals. However, due to the random nature of water molecule diffusion, it is difficult to form a regular layer of water molecules at the interface, which leads to failure of the adhesion work.

Keywords: Pavement engineering; rubberised asphalt; molecular dynamics; foaming mechanism; adhesion performance

* Corresponding author: Jiupeng Zhang, Jiupengzhang@chd.edu.cn