Microscopically analyzed interface behavior characteristics of acid precipitation on asphalt surface

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ABSTRACT

It is generally believed that the interaction between acid precipitation and asphalt has an effect on the performance of drainage asphalt pavement, but studies on the interface behavior characteristics from microscopic analysis are rare. Therefore, molecular dynamics (MD) was used to simulate the interfacial transition zone of precipitation (neutral and acid) and asphalt in the study, and the interfacial behavior characteristics of precipitation on asphalt surface were microscopically analyzed. Additionally, the composition of acid precipitation was configured in the laboratory, and the contact angles of precipitation solutions (SO$_4^{2-}$/CO$_3^{2-}$ and NO$_3^{-}$/CO$_3^{2-}$) on asphalt surface also verified the interface behavior characteristics between acid precipitation and asphalt. The results showed that the interaction of acid precipitation and asphalt is stronger than that of neutral precipitation, which makes it more difficult to remove from the surface of drainage asphalt pavement. With the increase of service temperature for drainage asphalt pavement, the interaction energy increases. Under the coupling effect of acid precipitation and low service temperature, water damage to the drainage asphalt pavement is more easily induced. The results revealed the micro-effect of acid precipitation and service temperature on drainage efficiency of an asphalt surface, which has certain theoretical significance and practical value for the application and exploration of drainage asphalt pavement.

Key words: acid precipitation, drainage asphalt pavement, interface behavior characteristics, molecular simulation, Sponge City

HIGHLIGHTS

- Interaction energy between acid precipitation and asphalt is stronger.
- As the service temperature rises, the interface behavior of precipitation on an asphalt surface becomes more visibly active.
- The coupling of acid precipitation and low temperature will further increase the influence of drainage asphalt pavement.
- Acid precipitation has a smaller contact angle, that is, better wettability.

1. INTRODUCTION

With the continuous industrialization of all countries in the world, the natural environment has inevitably been profoundly affected, among which the pH value (hydrogen ion concentration) of precipitation is the most easily perceived (Zhao et al. 2017; Zhou et al. 2019; Galella et al. 2021). Since coal and petroleum fuels have been used, the levels of acidic chemicals in acid precipitation has increased (the major acid precipitation areas in the world are shown in Figure 1), which is contrary to the concept of Sponge City that promotes sustainable development (Stempihar et al. 2012; Weiss et al. 2019). The appearance of acid precipitation not only affects the performance of drainage asphalt pavement in Sponge City, but also harms the durability of the asphalt surface (Chen et al. 2020; Guan et al. 2021). Therefore, it is very necessary to study the relationship between precipitation properties and the asphalt surface for the sustainable service performance of drainage asphalt pavement.

It is obvious from Figure 1 (https://neo.sci.gsfc.nasa.gov/) that acid precipitation is mainly distributed in Europe, North America and East Asia, where the world's major cities and populations are concentrated. Coincidentally, areas of low temperature (January) have a similar distribution to acid precipitation areas (Gao et al. 2021). Under the coupling effect of acid precipitation and low service temperature, the working performance of drainage asphalt pavement is inevitably affected...
(Lee et al. 2012). Obviously, this influence has a significant impact on the working efficiency and sustainable service performance of drainage asphalt pavement in Sponge City.

Traditionally, the influence of precipitation on drainage asphalt pavement is mainly reflected in water damage. For example, the performance of drainage asphalt pavement changes during precipitation (Guo et al. 2019), the optimization of pavement structure design for drainage asphalt pavement in Sponge City was beneficial to drainage (Lee et al. 2011), after comparison and selection of pavement drainage performance at low temperature (Collins et al. 2008). In addition, when the change of service temperature for drainage asphalt pavement was considered, the coupling effect of low temperature and precipitation becomes an important factor affecting the asphalt mixture (Tan et al. 2010; Xiao 2010). With the advent of acid precipitation, Roseen investigated the ability of drainage asphalt pavements to respond to different water and temperature characteristics, Zhang focused on the analysis of the performance of asphalt mixture under the influence of acidic precipitation (Zhang et al. 2004, 2005; Roseen et al. 2012). However, macroexperimental studies and performance tests can only seek rules and summaries from the changes of evaluation indexes, without essentially explaining their causes. Fortunately, molecular simulation provides a method for studying the interface interaction between precipitation and asphalt, and provides a new direction for microresearch (Ding et al. 2016; Yu et al. 2020a, 2020b; Zhang et al. 2021).

In conclusion, studies on the relationship between service environment (precipitation and temperature) and drainage asphalt pavement have greatly improved its service performance and durability. However, under the influence of temperature, the interface behavior characteristics of acid precipitation on asphalt surface have not been paid attention. Thus, a molecular dynamics method was used to simulate the interaction of acid precipitation on an asphalt surface. Combined with the change of service temperature, the interface behavior characteristics were microscopically analyzed. At the same time, the rationality of simulation was also verified by macroexperimental results.

2. MICRO-EXPRESSION OF PRECIPITATION AND ASPHALT

2.1. Precipitation with different pH value and temperatures

In the process of human production activities, the neutral state of precipitation in nature is broken. Acid precipitation (pH < 5.6) reminds us of the severity of environmental change. After analysis, acid precipitation contains sulfuric acid, nitric acid and hydrochloric acid, etc., which are easily completely ionized in water (Overrein 1983). This is mainly caused by the combustion of fossil fuels, so SO$_4^{2-}$ and NO$_3^-$ are the two main ions in acid precipitation (Askins 2007).

In addition, due to the influence of service temperature, the morphology of precipitation on asphalt surfaces will also change (as shown in Figure 2). Especially in cold regions, the drop in temperature causes precipitation to freeze. This directly increases the removal of precipitation from asphalt surfaces, which is not conducive to the service and durability of drainage asphalt pavement (Soper 2002). For example, under normal temperature environments, the precipitation on the road surface can be quickly eliminated, but the influence of rain and snow on the road surface is greater under low temperature.
2.2. Micro-expression of asphalt and precipitation

The four-component method (asphaltene, resin, saturation and aromatic) is generally considered to be a reasonable method to classify complex and diverse asphalt (Cui et al. 2020). Therefore, the asphalt four-component method was used in this study to characterize the microstructure. The proportions and chemical formula of the four components were determined through tests, and the calculation results are shown in Table 1 (Wang et al. 2017; Chu et al. 2019).

To define the proportion for asphalt components, molecular dynamics software was used to add and form an asphalt micro model. The asphalt formation model is shown in Figure 3.

In addition, micro models of precipitation can also be established by molecular simulation software. When precipitation is acid precipitation, it contains sulfate and nitrate ions. In addition, the cryogenic water will be frozen into ice crystals, as shown in Figure 4.

In order to verify the rationality of the micro models, simulated annealing was performed on the models. The energy of the system decreases reasonably with the simulation, and finally reaches the lowest state, that is, the stable state. On this basis, cohesive energy density (CED) and solubility parameters (SP) were calculated for the microscopic models. After comparison with the reasonable data range, the reasonableness of the model was determined (Shen et al. 2016).

2.3. Assembly and simulation of asphalt and precipitation molecules

The asphalt micro model and precipitation micro models verified by rationality were combined into a model, as shown in Figure 5. In the combined micro models, it can be divided into a vacuum layer, precipitation layer and asphalt layer. It is

| Components | Asphaltene | Resin | Saturation | Aromatic | Density/(g/cm³) |
|------------|------------|-------|------------|----------|----------------|
| Chemical formula | C₅₄H₆₅NOS | C₁₀₂H₁₁₀ | C₂₂H₄₆ | C₁₂H₁₂ | – |
| Percentage | 5.66% | 18.16% | 37.32% | 31.32% | 0.96 |
| Molecular number | 1 | 2 | 33 | 14 | |

Figure 3 | The 3D molecular lattice of asphalt.
found that although the asphalt layer is the same, the precipitation layer has different states and components. Thus, the interface behavior characteristics of precipitation on asphalt surface will be different.

The assembly models were built in preparation for the molecular dynamics simulation, and geometric optimization before running the simulation was beneficial to improve the simulation efficiency. Asphalt is a cementitious matrix composite material, which can be described by polymer field, while precipitation can be described by SPC field (Mullaney & Lucke 2014; Murashima et al. 2019). Dynamic simulations of asphalt and precipitation (ice crystals) were performed at 248 and 273 K at negative temperatures, respectively. The dynamic simulation of asphalt and precipitation was carried out at 273, 298 and 323 K, respectively, at positive temperature. After the system was simulated and analyzed by microscopic Newtonian forces, samples were extracted from the system, and then the properties of the whole system were calculated.

In summary, the data query and study proved the reliability of the selection for the four components. The state of different precipitation at different temperatures is also close to the actual state. This provides a good guarantee for the accuracy and rationality of simulation.
3. MICRO-ANALYSIS OF THE INTERFACE BEHAVIOR CHARACTERISTICS FOR PRECIPITATION ON ASPHALT SURFACE

3.1. Definition of interaction energy

According to material surface and interface science, when different materials come into contact with each other, interaction is inevitable. This is due to the composition of the different atoms, the two interact with each other to form an interface transition zone. Thus, the interface behavior characteristics of precipitation on an asphalt surface is inevitably affected by the precipitation properties (Jurečka et al. 2006; Sun et al. 2018). The interaction energy can be calculated by the energy difference between the sum of the whole and the parts. For example, the interaction energy of precipitation and asphalt \( (E_{\text{interaction}}) \) can be expressed in Equation 1, the total energy (precipitation and asphalt) is \( E_{\text{total}} \), the steady state energy of asphalt is \( E_{\text{asphalt}} \) and the steady state energy of precipitation is \( E_{\text{precipitation}} \):

\[
E_{\text{interaction}} = E_{\text{total}} - (E_{\text{asphalt}} + E_{\text{precipitation}})
\]  

(1)

3.2. Interaction energy between precipitation and asphalt

According to the method of molecular dynamics simulation, the interaction between acid precipitation (\( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \)) and asphalt was simulated at 0 °C (precipitation–asphalt), 25 °C (precipitation–asphalt) and 50 °C (precipitation–asphalt), respectively. The change in interaction energy at different temperatures is shown in Table 2 (according to Equation 1).

| Temperature | Acid precipitation (\( \text{SO}_4^{2-} \)) | Acid precipitation (\( \text{NO}_3^- \)) |
|-------------|------------------|------------------|
| 0 °C        | \(-2977.29\)     | \(-2689.82\)     |
| 25 °C       | \(-2436.37\)     | \(-2081.75\)     |
| 50 °C       | \(-1882.70\)     | \(-1531.33\)     |

It can be seen from Table 2 and Figure 6 that the values of interaction energy are different at different temperatures, but they are all negative. According to the definition of interaction, when the interaction energy is negative, there is mutual attraction between two materials. In addition, with the increase of temperature, the interaction energy gradually increases, which is not conducive to the removal of acid precipitation on the asphalt surface. But the high temperature is conducive to evaporation of acid precipitation.

When low temperature is considered further, the freezing phenomenon is presented. The simulation results of the interaction energy between precipitation and asphalt at this time are shown in Figure 7. The simulation was carried out at

Table 2 | Simulation results of interaction energy for acid precipitation (pH < 7)

| Temperature | Acid precipitation (\( \text{SO}_4^{2-} \)) | Acid precipitation (\( \text{NO}_3^- \)) |
|-------------|------------------|------------------|
| 0 °C        | \(-5852.72\)     | \(-5530.56\)     |
| 25 °C       | \(-5666.12\)     | \(-5318.71\)     |
| 50 °C       | \(-5427.70\)     | \(-5118.12\)     |

Figure 6 | Interaction energy between acid precipitation and asphalt.
-25 °C (ice-asphalt), -0 °C (ice-asphalt), 0 °C (precipitation-asphalt), 25 °C(precipitation-asphalt) and 50 °C (precipitation-asphalt).

It is obvious from Figure 7 that acid precipitation has more interaction energy than neutral precipitation. The interaction between surface acid precipitation and asphalt is stronger, which is more unfavorable for drainage of precipitation from asphalt surface. When low temperature is taken into account, solid precipitation is not conducive to drainage, although the interaction energy is small. Especially when acid precipitation and low temperature exist simultaneously, drainage asphalt pavement not only has to face the predicament of drainage function, but also has to endure the erosion of acid precipitation.

To sum up, the interaction between precipitation and asphalt is attractive, which is detrimental to the drainage function for drainage asphalt pavement. Although the interaction energy is minimal below 0 °C, this is of no significance for drainage function. At the same time, acid precipitation cannot be excluded, when low temperature freezing is superimposed, drainage asphalt pavement will face greater challenges. The specific analysis of the interface behavior characteristics of acid precipitation on the asphalt surface needs to be further studied.

3.3. Interface behavior characteristics of precipitation on asphalt surface

Because of the difference between acid precipitation and neutral precipitation, the interface behavior characteristics on asphalt surface are different. At the same temperature, details of the interaction of different precipitation on asphalt surfaces are shown in Figure 8.

From the micro details in Figure 8, it can be intuitively found that neutral precipitation is more regular on the asphalt surface. Meanwhile, acid precipitation is more active on the asphalt surface. Because of the SO$_4^{2-}$ and NO$_3^-$, acid precipitation and asphalt surfaces contact more closely. This results in more contact time between acid precipitation and asphalt at the macro level. Then the sustainable service performance of drainage asphalt pavement is challenged. In addition, the number and size of gaps between neutral precipitation and asphalt surface increase obviously. In the interfacial behavior of acid precipitation (SO$_4^{2-}$) and acid precipitation (NO$_3^-$) on asphalt surfaces, acid precipitation (SO$_4^{2-}$) is more stable. It can also be seen from the dispersion degree of water molecules outside the lattice that acid precipitation has a more active behavior, and the interaction between precipitation and asphalt can be enhanced.

When the temperature drops, precipitation inevitably freezes, and form ice crystals on a microscopic level as shown in Figure 9. The interface between precipitation (ice crystals) and asphalt has special characteristics.

As shown in Figure 9, in the simulation results of ice crystals and asphalt at 0 °C, the ice crystals at interface transition zone are affected by the asphalt surface without ice crystal morphology. The gaps between ice crystals and asphalt are larger, and the asphalt surface is visually rough. Although the interaction energy between ice crystals and asphalt is small, ice crystals are solid, so the drainage effect is almost nonexistent. However, if the ice crystals formed by acid precipitation are bound to increase the damage of asphalt surface, when freezing–thawing occurs, the residual water aggravates the water damage. Thus, although ice crystals have little significance for drainage function, the influence of acid precipitation on drainage asphalt pavement happens all the time.

In conclusion, the interaction energy (attraction) of acid precipitation on asphalt surface is stronger than that of neutral precipitation. The behavior of SO$_4^{2-}$ and NO$_3^-$ in acid precipitation on asphalt surfaces is similar. With the increase of
temperature, the interaction energy between precipitation and asphalt is enhanced, which is not beneficial to drainage. Although freezing at low temperature has little significance for drainage, it is disadvantageous in terms of water damage. Meanwhile, with the coupling of low temperature and acid precipitation, the durability and function of drainage asphalt pavement are greatly challenged.

**Figure 8** | Interface behavior characteristics of precipitation on an asphalt surface. (a) Interfacial behavior of precipitation on an asphalt surface (25 °C). (b) Interfacial behavior of acid precipitation on an asphalt surface (25 °C SO$_4^{2-}$). (c) Interfacial behavior of acid precipitation on an asphalt surface (25 °C, NO$_3^-$).

**Figure 9** | Interfacial behavior of ice crystals on an asphalt surface.
4. VALIDATION OF DRAINAGE ASPHALT PAVEMENT COUPLED WITH ACID PRECIPITATION AND LOW TEMPERATURE AND ITS INFLUENCE ON SPONGE CITY

4.1. Contact analysis of acid precipitation on asphalt surface

In order to verify the accuracy of simulation results, macro experiments were necessary. So sulfuric acid solution and nitric acid solution were formulated in the laboratory to simulate acid precipitation. The pH values of neutral precipitation and acid precipitation (SO$_4^{2-}$ and NO$_3^-$) solutions are shown in Figure 10.

As shown in Figure 10, the three solutions represent different precipitation types. Traditionally, the pH values of acid precipitation is less than 5.6. In order to increase the effect of contact angle experiments, the pH values of acid solutions were increased. In addition, asphalt 90# was evenly applied to the glass sheet (Sprinz 1992; Yu et al. 2020a, 2020b). After the natural precipitation was simulated in the laboratory, contact angle experiments were used to observe the contact of precipitation on the asphalt surface, as shown in Figure 11. In order to increase the authenticity of precipitation, a dropper was used as precipitation droplet formation. The image was processed and the contact angles on the left and right were calculated. The average values are shown in Table 3.

From the change of contact angle, it can be confirmed that the contact angle of neutral precipitation is obviously larger than that of acid precipitation. This indicates that the wettability of asphalt is poor by neutral precipitation, while the wettability of acid precipitation is stronger on the asphalt surface. In addition, a sulfate solution has more obvious wettability than a nitrate solution. Therefore, acid precipitation has stronger wettability on an asphalt surface, and it is more difficult to exclude acid precipitation on drainage asphalt pavement. This poses great challenges to the functional and structural aspects of drainage asphalt pavement.

4.2. Influence of coupling of acid precipitation and low temperature on drainage asphalt pavement

In urban construction, the main road covers 10% to 15% of the ground area, which can reach 30% to 40% in developed cities (Li et al. 2017; Xia et al. 2017). At the same time, in order to delay the peak runoff and water resources recycling, drainage asphalt pavement provides a good method for urban road rainwater collection. In Figure 12 shows drainage asphalt pavement in Sponge City.

![Figure 10](image1.png)  | Precipitation solutions with different pH values.

![Figure 11](image2.png)  | Contact angle of precipitation solutions.
Unfortunately, from the global temperature distribution, it can be seen that the extent of seasonal freeze is similar to urban distribution (Figure 13). When low temperature and acid precipitation occur simultaneously, it is a great challenge to the construction and operation of Sponge City. Therefore, the structure and function of drainage asphalt pavement are threatened by acid precipitation and low temperature environment. The normal operation of Sponge City requires good drainage of asphalt, while acid precipitation requires global efforts to protect the environment and reduce pollution.

In conclusion, the study reveals the interaction mechanism between acid precipitation and asphalt from the microscopic point of view, and provides a theoretical basis for the study of acid precipitation damage for drainage asphalt pavement. Combined with the influence of service temperature, it provides a new research direction for the development of drainage asphalt pavement in freezing–thawing areas. This lays a foundation for promoting the sustainable development of Sponge City and is conducive to further improving the utilization rate of water resources.

5. CONCLUSIONS

(1) Interaction energy between acid precipitation and asphalt is stronger than that of neutral precipitation, which is more unfavorable to the removal of precipitation from drainage asphalt pavement in Sponge City.

(2) As the service temperature rises, the interface behavior of precipitation on the asphalt surface becomes more visibly active and has stronger interaction energy.

(3) The coupling of acid precipitation and low temperature will further increase the influence of acid precipitation on the structure and function for drainage asphalt pavement.

(4) Hydrophilicity of acid precipitation was also verified by the contact angle experiments. An acid precipitation solution has a smaller contact angle, that is, better wettability. At the moment, acid precipitation is more difficult to drain, which challenges the durability and functionality of drainage asphalt pavement.

### Table 3 | Contact angle between precipitation and asphalt

| Contact angle | Precipitation | Acid precipitation (SO$_4^{2-}$) | Acid precipitation (NO$_3^-$) |
|---------------|---------------|----------------------------------|-------------------------------|
| Left          | 72.444        | 61.700                           | 70.370                        |
| Right         | 74.868        | 63.165                           | 69.870                        |
| Contact angle | 73.656        | 62.432                           | 70.120                        |

### Figure 12 | Drainage asphalt pavement in Sponge City construction.

### Figure 13 | Global average surface temperature distribution (https://Neo.Sci.Gsfc.Nasa.Gov/).
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DISCLOSURE STATEMENT

No potential conflict of interest was reported by the authors.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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