Interface Bonding Mechanism of Recycled Asphalt Mixture

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Abstract: With more and more asphalt pavement entering into maintenance period, a large number of aged and damaged asphalt pavements are excavated to reclaimed asphalt pavement. Recycling of old asphalt pavement into asphalt mixture is one of the important ways to solve this kind of waste material. However, the water stability of reclaimed asphalt mixture is difficult to meet the requirements of actual pavement performance, resulting in insufficient pavement durability. Water vapor weakens the bond between asphalt and aggregate and asphalt paste. In this paper, the failure mechanism of interfacial bond in asphalt mixture is summarized by using weak boundary layer, electrostatic force, chemical bond, mechanical bonding and surface free energy from the view of microstructure. Further more, the research direction of interfacial bonding mechanism of recycled asphalt mixture is put forward.

1. Introduction
The performance of asphalt pavement is degraded over long-term service, and it is easy to be damaged under the action of driving load. In order to guarantee the function of asphalt pavement, it is necessary to carry on milling or digging out the damaged pavement. As a large number of asphalt surfaces enter the maintenance period, more and more old asphalt pavement materials will be produced.

The United States produces 300 million tons of reclaimed asphalt pavement materials every year, and Japan reaches 50 million tons. At present, China is over 90 million tons and is growing at a rapid rate of 15% a year [1]. In order to reduce the mining of the new aggregate, protect the natural environment, restrain and solve the problems such as the pollution of the old pavement material and the land occupied by the reclaimed asphalt pavement, realizing the economy of recycling and utilization of the old asphalt pavement [2], it is imperative to recycle the old asphalt pavement into asphalt mixture.

2. Water stability of recycled asphalt mixture
One of the main reasons for the degradation of asphalt pavement material performance is water damage, which is usually manifested in pavement deformation, loose, cracking, asphalt peeling and aggregate fly-off and other types [3,4]. The moisture in asphalt mixture weakens the bonding action between asphalt and aggregate, and the bonding effect between asphalt mortars in microstructure, and there are fine cracks in the ineffectual part of the asphalt mixture, and the water content in the asphalt mixture weakens the bonding action between asphalt and aggregate. The crack propagation of asphalt mixture can occur either at the binder-aggregate interface or within the binder. The former is called adhesive cracking, and the latter is called cohesive cracking. Generally, moisture in the liquid or steam state weakens the bonding between asphalt and aggregate and asphalt paste, and moisture will affect the...
microstructure of the material. Finally, it leads to the degradation of the macro-structural mechanical properties of asphalt pavement mixture [5], which has undergone several physical and chemical processes, and its mechanism is not fully understood.

The recycled material of asphalt pavement is different from ordinary aggregates. The recycling material of asphalt pavement is studied by using the industrial CT of X-ray tomography technique in my doctoral thesis, and it is found that it is a kind of coarse and fine aggregate complex. From the microcosmic point of view, it is found that the particle shape of asphalt pavement recycled material is quite different from that of ordinary aggregates, because of the effect of asphalt aging and water invasion. There are some problems such as the bonding between asphalt and aggregate and the weak bonding between asphalt grout and so on. The water stability of asphalt mixture produced by asphalt pavement recycled material is slightly lower than that of new aggregate mixture, and the water stability of asphalt mixture produced by asphalt pavement recycling material is less stable than that of new aggregate mixture. Many researches have adopted recycled asphalt mixture with recycled asphalt pavement. The results of indoor performance test and road performance study show that the water stability is difficult to meet the practical requirements of service performance for road [7-9]. Therefore, water stability is the key to restrict the durability of pavement using recycled asphalt mixture. Moisture weakens the bond between asphalt and aggregate, and the internal bond failure mechanism of recycled asphalt mixture from the view of microstructure need to be studied. Understanding the degradation principle of adhesive performance of recycled Asphalt mixture is an important research direction to solve the problem of water stability for pavement.

3. Interfacial bonding mechanism of asphalt mixture

The interfacial adhesion between asphalt and aggregate had been studied by dynamic rheometer, differential scanning calorimetry (DSC), atomic force microscopy (AFM), liquid chromatography (HPLC). Centrifugal and infrared spectroscopy techniques were used to study the adhesion properties of water vapor to the interface between asphalt and aggregate at the level of microstructure [10]. In general, the following five theories have better explained the bonding properties of microstructure of asphalt mixture. They are weak boundary layer, electrostatic force, chemical bond, mechanical bonding, surface free energy and so on.

**Weak boundary layer**

It is pointed out that the bond failure may be due to the low adhesion strength in the interface regions [11]. If the aggregate containing dust on the surface is not effective in bonding with asphalt, it is due to the formation of weak boundary layer between dust and asphalt. The thickness of the asphalt film affects the interfacial adhesion property. If the asphalt film is very thin, the adhesion cracking between the asphalt and the aggregate will occur, and if the asphalt film is thicker, the asphalt paste adhesion cracking will occur [12], and compared with the adhesion effect, the adhesion cracking of asphalt paste will occur when the asphalt film is very thin. Adhesion plays a more important role in the destruction of water vapor by stripping asphalt from the aggregate surface.

**Electrostatic force**

The electrostatic force theory ascribes the bond strength between asphalt and aggregate to the ionic Coulomb gravity on the surface of the material [13]. When the surface of asphalt and aggregate permeates with water vapor, it forms an electric layer, which is divided into the inner layer and the outer layer. The inner layer is composed of one or more layers of ions with opposite charges, and the outer layer is moved and diffused by the inner layer ions. When pH and EMF of aqueous solution have the same polarity (such as acid aggregate), this will lead to net repulsion between asphalt and aggregate and remove asphalt from aggregate surface.

**Chemical bond**

Cohesion between asphalt and aggregate is the result of chemical reaction between the two materials. The chemical bond is an interface material formed by the reaction between active functional group and aggregate in asphalt. Hefer et al explained how different chemical active sites on the surface of mineral aggregates react with various functional groups in bitumen binders, resulting in interfacial adhesion between the two materials [14]. Generally speaking, aggregates with high carbonate content (also known as alkaline aggregates) are more likely to be coated with binders than those with high silicon content (also known as acid aggregates). This is because siliceous aggregates contain high
concentrations of hydroxyl groups, which have greater affinity with carboxylic acids and water. The carboxylic acid components in the binder are adsorbed on the surface of these aggregates to form asphalt-aggregate bonds. However, in the presence of water, the bond with carboxylic acid is also prone to shift. The chemical composition and mineral composition of aggregate determine the quality and durability of asphalt binder. Adhesion due to chemical bonds also helps to explain the role of hydration lime and anti-stripping additives in improving the moisture resistance of asphalt mixtures [15]. They also show that the stress relief ability of asphalt binder and the fatigue damage resistance of the mixture are improved.

**Surface free energy** Surface free energy refers to the increment of the corresponding thermodynamic function when the unit surface area is increased by keeping the corresponding characteristic variables unchanged. The physical and chemical adhesion between asphalt and aggregate can be seen as a thermodynamic phenomenon, which requires the removal of asphalt from the aggregate surface (bond failure), that is, the work or energy required to produce new units of area in a vacuum. The physical and chemical adhesion between asphalt and aggregate depends on the surface free energy of the material. The higher surface free energy (adhesion work) indicates that the adhesion strength between asphalt and aggregate interface is better [16].

4. **Application of surface free energy**
According to the binding energy between asphalt and aggregate and the change of surface free energy when water replaces asphalt from aggregate surface, the degree of water damage of asphalt mixture can be quantified. The process of asphalt mixture being damaged by water is explained by thermodynamic theory as a spontaneous process. The larger the binding energy between asphalt and aggregate is, the smaller the value of free energy decrease in the system under the influence of water, under the condition of drying without water vapor, the higher the binding energy between asphalt and aggregate is, the less the value of free energy in the system is affected by water. The more favorable asphalt mixture is to restrain water damage. The surface free energy theory provides a good method to explain the water damage of asphalt mixture, which can be used as an important thermodynamic parameter in the damage constitutive model of asphalt mixture. It also provides the possibility of selecting aggregate and asphalt in the design of improving the water damage resistance of asphalt mixture. However, the study on surface free energy theory of recycled asphalt mixture is rarely reported. From the microscopic point of view, it is found that the particle surface morphology of asphalt pavement recycled materials is quite different from that of ordinary aggregates, and the surface free energy of the materials may vary greatly compared with common aggregates and bitumen. Therefore, from the point of view of microstructure, The adhesive strength of recycled asphalt mixture is interpreted by the theory of interfacial adhesion between asphalt and aggregate, which can improve the water stability of recycled asphalt mixture and point out the research direction to solve the water damage of recycled asphalt pavement.

The adhesion between asphalt and aggregate mainly depends on the physical and chemical adhesion caused by the free energy of the surface of the material and the mechanical interlock between the machines. The mechanical interlocking action also depends mainly on the bond strength and / or bonding strength caused by the free energy of the surface of the material or the chemical bonding between the materials [19]. Reference [20] has proved that the viscosity reduction of asphalt mixture caused by water damage is closely related to the change of free energy on the surface of asphalt mixture, and the decrease of free energy is also shown to be related to the change of free energy of asphalt binder. A function between aggregates and free energy components on the surface of water were deduced. Reference [21] established a model based on the surface free energy of asphalt adhesive and aggregate, and successfully predicted the effect of admixture on improving the water stability of asphalt mixture. The relationship between the energy parameters based on surface free energy and the water stability of asphalt mixture is also discussed. However, these studies all consider the common aggregate and asphalt. There are some asphalt binder wetting or coated aggregate surface in the aging asphalt mixture, and the surface free energy is obviously different from the ordinary aggregate. Based on the in-depth study of the surface free energy of the aged asphalt mixture, the comparison of the difference with the common aggregate,
combined with the prediction model of the water stability of the common asphalt concrete, it is expected to find out the bonding mechanism of the microstructure of the recycled asphalt mixture.

5. Conclusion

The insufficient water stability of recycled asphalt mixture is an urgent problem to be solved in the application of recycled asphalt mixture. In the previous research, it was found that the surface free energy theory can better explain the water stability of asphalt mixture from the micro point of view. The research of the author's doctoral thesis found that the particle surface morphology of asphalt pavement recycled material is different from that of ordinary aggregate, and the phenomenon of asphalt aging and peeling aggregate surface has already occurred in some interface regions. Therefore, this study proposed that the particle surface of asphalt pavement recycled material is different from that of ordinary aggregates. The variation of surface free energy of recycled asphalt pavement results in insufficient water stability of recycled asphalt mixture. On this basis, the surface free energy of different components of recycled asphalt mixture need to be studied and tested. Furthermore, the water stability of recycled asphalt mixture can be improved by improving the surface free energy of each component.

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