Construction and Building Materials 132 (2017) 230–239

Contents lists available at ScienceDirect

Construction and Building Materials

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

Identification of wetting and molecular diffusion stages during self-healing process of asphalt binder via fluorescence microscope

Daquan Sun, Guoqiang Sun, Xingyi Zhu *, Qi Pang, Fan Yu, Tianban Lin

Key Laboratory of Road and Traffic Engineering of Ministry of Education, Tongji University, Shanghai 200092, PR China

HIGHLIGHTS

- The Two-Stage Model was built to characterize the healing process.
- SBS modified asphalt shows larger regain strength and greater diffusion healing rate.
- Strength is not completely recovered as though the crack has fully closed.

ARTICLE INFO

Article history:
Received 30 July 2016
Received in revised form 18 October 2016
Accepted 30 November 2016

Keywords:
Asphalt
Self-healing
Two-Stage Model
Wetting healing
Molecular diffusion healing

ABSTRACT

In order to distinguish the wetting healing stage and the molecular diffusion healing stage during the self-healing process of asphalt binders, the fluorescence microscope (FM) together with the image processing technique was employed. The healing index was put forward based on the variation of crack area. On the basis of the established capillary diffusion theory, the Two-Stage Model was built to fit the healing curves characterizing the healing index variation with healing time. It is found that the established method is quite suitable to identify the wetting and the diffusion stages. The wetting healing occurs firstly, during which the asphalt molecules spontaneously wet the both faces of crack. Although the strength recovery is not distinct at this stage, SBS modified asphalt shows greater wetting healing strength than the other neat asphalt binders. Once the wetting process is nearly finished, the molecular diffusion healing ratio accelerates significantly, and the descending order of the molecular diffusion rate is: SBS > PEN 100 > PEN 70 > PEN 50 > PEN 20. The test results also show that the disappearance of visible crack does not indicate that the strength has been fully recovered. Here, the tensile strength test is developed to identify the strength recovery rate after the crack is fully closed, which implies that the strength regeneration is a long process.

1. Introduction

The failure process of composites is normally preceded by the coalescence of interacting micro cracks to form a fatal crack. The formation of micro cracks with a length on the order of a grain size is always regarded as the initial step of fracture development. Therefore, if all the micro cracks are healed as early as possible, the service life of composites can be greatly prolonged.

Self-healing plays a vital role in extending the fatigue life of asphalt materials [1–3]. Since the self-healing phenomenon in asphalt mixture was first reported in 1960s [4], it has become a hot research field. Among the early studies, most researchers investigated the healing properties of asphalt materials by adding a certain interval time during the repeated fatigue loading period. Then, a series of indicators—strength, energy, and fatigue life recovery ratios were employed to evaluate the self-healing capability [5–10]. Hence, it is critical to establish a reasonable self-healing model and analyze the healing process based on a proper test.

Many researchers paid attention to the self-healing mechanism of asphalt materials and proposed different self-healing models based on different theories. According to the fracture surface energy theory, Little and Lytton et al. [11–13] established the self-healing equation of asphalt concrete, which characterized the relationship between the healing rate and the surface energy of fracture. Wool and Kim et al. [14–17] discussed the healing mechanism of polymer materials and built a diffusion model which divided the healing of the polymer into the wetting healing in crack face and the intrinsic healing of the material. Based on the theory of Wool et al., Bhasin and Little [18,19] conceived that the short-term self-healing of asphalt was related with the wetting

* Corresponding author.
E-mail address: zhuxingyi66@aliyun.com (X. Zhu).

http://dx.doi.org/10.1016/j.conbuildmat.2016.11.137
0950-0618/

mechanism, while the long-term self-healing was ascribed to the diffusion mechanism. Álvaro García [20] further analyzed the asphalt healing process combining with the capillary mechanical theory and then built the capillary flow model. The corresponding hypothesis was put forward: under the proper temperature condition, the capillary flow in the micro crack of the asphalt caused the crack closure and the interface strength was gradually recovered due to the diffusion of the interface molecules. Following the work of Wool and Kim et al., Sun et al. gave the self-healing equation of asphalt binder and developed a macroscopic recovery function of asphalt materials based on the Arrhenius law together with the fatigue-rest-fatigue test [21]. Yet the fatigue crack size is too small to be measured by the conventional instrument. Recently, Qiu et al. [22–24] recorded the dynamic micro crack healing process of the neat asphalt and the SBS modified asphalt with the aid of the fluorescence microscope (FM) which can clearly keep a record of the healing process.

The valuable works above deepened the understanding of the self-healing mechanism of asphalt materials from the view of molecules and energy. However, the healing process (including the surface approach, wetting, molecular diffusion, and randomization) is very difficult to be precisely detected and characterized at micro-scale [15,18,19]. Finding the appropriate detection technology is the key to describe the healing process accurately.

In this paper, a Two-Stage Model based on the capillary diffusion theory was established to analyze the wetting healing process and the molecular diffusion healing process of various asphalt binders. The healing process of fractured asphalt specimens was observed by FM to monitor the real time change of the crack area from the meso scale. Together with the image processing technique, the healing index based on the change of the crack area was put forward. By fitting the healing curves with the Two-Stage Model, the wetting healing stage and the molecular diffusion stage were identified. Then, the two healing stages were discussed to evaluate the healing efficiency of different types of asphalt binders including the neat asphalt and the SBS modified asphalt. Finally, the initial tensile strength and the healed tensile strength were measured by the force ductility test to distinguish the healing efficiency of various asphalt binders.

2. Methodology

Some researchers have pointed out that the self-healing process can be described by capillary diffusion theory [20–22,25,26]. If the asphalt or polymer-modified asphalt is heated to its glass transition temperature, it will liquefy, flowing sluggishly over long time periods. When encountering one of the propagating cracks including the contact points and the empty space, this liquefied asphalt enters the cracked face driven by capillary force [27]. After that, it may act as the healing agent, wetting the cracked region, diffusing into the fractured matrix driven by gradients of pressure or of concentration, and entangling with the matrix molecules by surface energy difference or thermodynamic driving force, see Fig. 1. Finally, the cracks disappear with the restoration of interfacial strength at a fracture surface. Actually, the capillary force does have a great effect on the healing efficiency, especially during the wetting healing stage. The capillary action reflects the ability of a liquid to flow in a thin tube or the porous materials without external assistance, or even with external resistance like gravity. When the diameter of the tube is small enough, the surface tension and adhesive forces between the liquid and container wall will act together to promote the liquid flow. According to the research of García et al. [20,26], the capillary force through the crack in asphalt is mainly related to the viscosity and surface energy of the asphalt binder as well as the temperature. They found the capillary flow driven by surface energy is the main cause of healing. Capillary action starts from these contact points between the both crack faces and then extends through the crack. As a result, a better healing effect will be achieved under a greater capillary force.

Based on the capillary diffusion theory above, the self-healing process can be mainly divided into two stages including wetting healing and molecular diffusion healing.

(1) Wetting healing stage

According to the Washburn capillary model [27], the crack can be simulated as a set of capillary tubes. Each tube is assumed to be circular and vertical for the purposes of calculation. For a given tube under constant capillary force (as shown at the wetting stage in Fig. 1), the volume of liquefied asphalt entering into the capillary tube is proportional to the square root of the wetting time t and the square root of the ratio of the surface tension to the viscosity γ/η [27], and it can be calculated as:

$$V = K(\gamma/\eta)^{0.5} \cdot t^{0.5} \tag{1}$$

where K is independent of the nature of asphalt and is related to the ambient temperature. The wetting healing strength can be obtained due to the cohesion of liquefied asphalt in crack faces driven by capillary force, so the wetting healing index can be expressed as:

$$R_0(T,t) = A \cdot t^{0.5} \tag{2}$$

where $R_0(T,t)$ is defined as the wetting index related to the ambient temperature T and the wetting time t. A is a constant which is related to the properties of the asphalt binder and the ambient temperature.

(2) Molecular diffusion healing stage

After a short-time wetting stage, the long term strength gain at the fracture surface mainly relies on the diffusion and randomization of the asphalt molecules. The diffusion healing strength is gradually formed due to the entanglement of the molecular chains and the diffusion of molecules. Wool and O'Conner [15] proposed the healing behavior equation of polymer, which has been proved to be suitable for asphalt materials [10,18,21,28–30] and is given by:

$$R(T,t) = R_0 + R_h(T,t) \tag{3}$$

where $R(T,t)$ represents the macroscopic healing index at temperature T and time t. R_0 represents the wetting healing strength gain of asphalt binder in a short period, and $R_h(T,t)$ represents time-dependent strength gain during the molecular diffusion stage and it can be expressed as:

$$R_h(T,t) = B \cdot t^n \tag{4}$$

where B is a temperature-dependent parameter, which indicates the strength gain rate due to the inter diffusion of molecules between the crack surfaces at temperature T [21]. The value of n is related to the molecular diffusion rate, and it is within 0.25–0.5.

It should be mentioned that the molecular diffusion explains the net flux of molecules from the region with higher concentration to one with lower concentration. Once the concentrations are equal, the molecules continue to move. However, if there is no concentration gradient, the diffusion process will stop and be instead governed by the process of self-diffusion, originating from the random molecular motion. That is to say, the molecular diffusion is a continuous process till the system reaches a dynamic equilibrium gradually. From the spatial and temporal scale, the outcome of the molecular diffusion can be detected through the meso-level.