



# Calculation and evaluation of activation energy as a self-healing indication of asphalt mastic



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## HIGHLIGHTS

- Healing function of asphalt materials based on Arrhenius law is developed. The standard test procedure to get activation energy is put forward.
- It proves that it is reasonable to adopt activation energy value as the indication to evaluate the self-healing ability of asphalt mastic.
- The effects of asphalt and filler, asphalt-filler ratio, damage degree on the activation energy and self-healing ability of asphalt mastic are discussed.

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## ABSTRACT

Self-healing, as a very valuable characteristic, should be considered when selecting and designing constitutions of asphalt materials. It is necessary to develop a fundamental and universal healing evaluation indication based on the healing mechanism. However, most existing healing evaluation indication lacks physical meaning and hardly can predict the time-dependent healing process under different conditions. In this paper, the formulation of activation energy as a self-healing indication is developed based on the Arrhenius law. The procedure to get the activation energy of different asphalt mastic is put forward. Nine samples of different asphalt mastic are prepared. Then, the fatigue–rest–fatigue tests are employed to get the activation energy of nine asphalt mastic. Finally, the potential of the activation energy as the healing evaluation index for asphalt mastic is studied, and the effect of type of asphalt and filler, asphalt-filler ratio, damage degree on the value of activation energy is discussed. It is found that the healing activation energy can obviously distinguish the healing ability of different asphalt mastic. Healing activation energy represents the minimum energy required for the time-dependent strength gain and reflects the time-dependent strength gain rate, also can rank the time-dependent healing capacity of different asphalt mastic.

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## 1. Introduction

Fatigue cracking is a common failure in asphalt pavement. The dominant mechanism in the fatigue damage process is microcracks development, growth and coalescence of microcracks, ultimately propagation in the form of macrocracks [1]. Theoretically, any material has a certain potential to heal the crack for reducing crack surface energy after crack occurred. As a viscous fluid, asphalt should have a relatively obvious healing behavior for its relatively strong surface wetting and diffusion capability. Since the first experimental demonstration of asphalt healing conducted by Deacon (1965), more laboratory investigations [2–5] and field observations [6,7] have clearly shown that asphalt binders and asphalt mixtures have the potential to heal cracks and recover part

of strength and stiffness when given a certain amount of resting time and sufficient energy. Self-healing, as a very valuable characteristic, should be considered when selecting and designing constitutions of asphalt materials. Hence, how to accurately and reasonably evaluate healing potential of asphalt materials becomes a top issue.

Most existing laboratory test methods to evaluate self-healing are empirical or phenomenological in nature, generally applying a continuous fatigue load to the sample until it gets a certain damage, and then allowing it to recover over a period of time. By comparing the performances (e.g. modulus and stiffness [8,9], fatigue life [10–12]) before and after the rest periods, several healing evaluation indexes are developed. These healing evaluation indexes can rank the healing capability of different asphalt materials under certain conditions, but the validity of these indexes depends on healing temperature and resting time to some extent. Some of indexes will be useless when resting time is long, healing

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temperature is high or damage introduced is limited. It is noted that all the existing evaluation indexes is comparable only at a given healing temperature and resting time. These indexes hardly predict the time-dependent healing process at the different healing temperature and perhaps will be questioned due to the lack of physical background.

To understand why the self-healing reactions would occur, we could reconsider it from the micro-scale point of view. When sufficient energy is transferred to the system (for example, the temperature of the system increases), the number of molecules that carry enough kinetic energy climb the activation energy barrier, wet the crack faces, diffuse from one face to the other when crack faces in rather close contact, and finally put both crack faces in contact [13]. Based on above self-healing behavior, the crucial issue is that how high the activation energy barrier is. The lower the activation energy barrier is, the less energy required for the molecules to begin the process of self-healing. Therefore, it seems reasonable to adopt the activation energy value as the indication to evaluate the self-healing ability of asphalt mastic. However, based on the knowledge of authors, seldom researches have been conducted to analyze the relationship between the activation energy and self-healing ability of asphalt mastic.

As we have known, asphalt mastic has a significant effect on the healing of asphalt mixtures because healing occurs either within asphalt mastic or at the interface between asphalt mastic and aggregate. However, limited research has paid attention to the healing of asphalt mastic. Therefore, the object of this study is to establish a healing evaluation index for asphalt mastic based on healing mechanisms. Since the observed macroscopic recovery can be considered as the contribution of the instantaneous strength gain and the time-dependent strength gain, a macroscopic recovery function of asphalt materials based on the Arrhenius law is developed firstly. The procedure to get the activation energy of different asphalt mastic is put forward. Nine samples of asphalt mastic with different type of asphalt and filler, asphalt-filler ratio, damage degree are prepared. Then, the fatigue–rest–fatigue tests are employed to investigate the self-healing ability of asphalt mastic. Finally, the potential of the activation energy as the healing evaluation index for asphalt mastic is studied, and the effect of asphalt type, filler type, asphalt-filler ratio, damage degree on the value of activation energy and self-healing ability is discussed.

## 2. Proposed methodology

In general, a healing process can be hypothesized to be the reverse process of cracking, which contains two important stages, namely, crack closure and strength gain [14]. Some micromechanical and phenomenological healing models in polymers have been proposed [15–18] to describe these two stages. One of the most well-known models is the interfacial healing model proposed by Wool and O'Connor [16–17], whose theory is based on molecular inter-diffusion. The model was developed by the convolution of an intrinsic healing function and wetting distribution function. Similar to the above self-healing processes, recent studies [2,19–21] pointed out the two primary healing processes for asphalt materials as well: (1) instantaneous strength is gained due to interfacial cohesion between the crack faces, and (2) long term strength is gained due to diffusion and randomization of molecules from one face to the other. Because of the similarities with the healing processes in asphalt mastic, and studies on asphalt materials that have successfully adopted the polymer models, the idea based on the previous work done by Wool and O'Connor is also being adopted in this study. Therefore, the observed macroscopic recovery at temperature  $T$  and time  $t$ ,  $R(T, t)$ , can be written as:

$$R(T, t) = R_0(T) + R_h(T, t) \quad (1)$$

where  $R_0(T)$  represents the instantaneous strength gain,  $R_h(T, t)$  represents time-dependent strength gain. Researchers [22–24] demonstrated that the polymer chain diffusion across interfaces forms the interface cohesive strength ( $R_h(T, t)$ ), which depends on the healing time to the power of 0.25. It is hypothesized that this relationship can be used into asphalt materials. Therefore, the observed macroscopic recovery function of asphalt materials can be rewritten as:

$$R(T, t) = R_0(T) + D(T)t^{0.25} \quad (2)$$

where  $D(T)$  is a temperature-dependent parameter, which indicates the strength gain rate due to the inter diffusion of molecules between the crack surfaces at temperature  $T$ , namely, diffusion rate.  $D(T)$  can be represented by the Arrhenius law of diffusion as defined in:

$$D(T) = D_0 \exp\left(-\frac{E_h}{RT}\right) \quad (3)$$

where  $D_0$  is a diffusion constant,  $R$  is the universal gas constant (8.314 J/mol/K),  $T$  is the temperature in degrees K.  $E_h$  is the activation energy for diffusion and randomization of molecules from one face to the other, which represents the minimum energy required for the time-dependent strength gain. In this study,  $E_h$  is also defined as the healing activation energy.

Substituting Eq. (3) into Eq. (2), the intrinsic healing function can be rewritten as:

$$R(T, t) = R_0(T) + D_0 \exp\left(-\frac{E_h}{RT}\right) t^{0.25} \quad (4)$$

Theoretically, Eq. (4) can be described as the healing process, as shown in Fig. 1.  $E_h$  is the healing activation energy, which varies with the type of asphalt and the type of damage introduced. If appreciable energy equal to or greater than the healing activation energy exists at the damage surface, the time-dependent strength gain will be initiated, namely, self-healing reaction will proceed. Otherwise, the reaction will be terminated. Therefore,  $E_h$  is a promising parameter to evaluate the self-healing potential of asphalt.

## 3. Experiments

### 3.1. Materials

Five representative asphalt binders were considered in this study, including neat asphalt (NA), SBS modified asphalt (SBS), rock asphalt modified asphalt (RA), warm mix asphalt (WMA), and high viscosity modified asphalt (SK).

The neat asphalt (penetration grade 60/80) was sampled from China Offshore Bitumen Company (Taizhou, China). SBS modified asphalt was prepared by mixing the neat asphalt and 5% (by weight of asphalt) SBS polymer at 175 °C for 60 min using a high speed shearing mixer. A rock asphalt modified asphalt was prepared by blending the neat asphalt and 8% (by weight of asphalt) Xingjing rock asphaltite at 160 °C for 30 min. Xingjing asphaltite was obtained from Xingjing Asphaltite Ltd. (Urumqi, China). The properties and composition of Xinjiang Asphaltite sample were reported in previous studies [25]. Warm mix asphalt was prepared by blending the neat asphalt and 0.7% (by weight of asphalt) Warm-Mix Additive (Rediset® LQ1106, Akzo Nobel Corporation) at 160 °C for 30 min. High viscosity modified asphalt was obtained from Korea SK Incheon Petrochem Company. The basic properties of these asphalt binders are listed in Table 1.

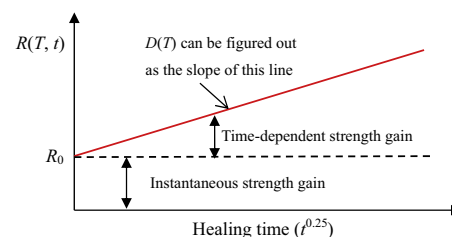


Fig. 1. Hypothesis of the healing process.

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