



## Novel vascular self-nourishing and self-healing hollow fibers containing oily rejuvenator for bitumen

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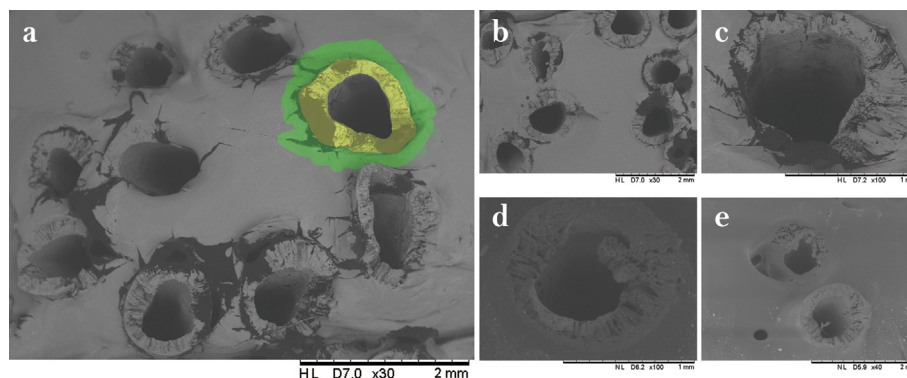
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### HIGHLIGHTS

- Novel self-nourishing concept was mentioned in this work.
- Hollow fibers containing oily rejuvenator were successfully fabricated.
- Self-nourishing and self-healing behaviors of rejuvenator were investigated in bitumen.

### GRAPHICAL ABSTRACT

ESEM morphologies of hollow fibers/bituminous material composites treated with various temperatures, (a) 180 °C, (b) 200 °C, (c) 220 °C, (d) 240 °C and (e) 260 °C.



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### ABSTRACT

The aim of this work was to prepare and characterize a novel vascular self-nourishing and self-healing hollow fiber containing oily rejuvenator for aged bitumen. The hollow fibers containing oily rejuvenator were fabricated with micropores by a one-step wet-spinning method using polyvinylidene fluoride (PVDF) resin. Hollow fibers had been successfully prepared based on the test results of morphology, size and chemical structure. The hollow fibers had a good thermal stability with a decomposition temperature of 450 °C. The mechanical properties of fibers satisfied the requirements of use in bituminous materials. Because of the existence of micropores microstructure in fibers, the rejuvenator had the permeability out of fibers. The diffusion behaviors of rejuvenator were also measured by an attenuated total reflection flouirier transformed infrared spectroscopy (FTIR-ATR) method. It was found that the diffusion coefficient greatly depended on the pore structure and temperature. A fitting model implied that the diffusion coefficient had a big deviation of Fick's law and greatly influenced by the pore structure of hollow fibers. It is a guide to design and regulate the microstructure of hollow fibers with appropriate rejuvenator releasing behaviors. All above results implied that the existence of both self-nourishing and self-healing had been successfully proved in aged bitumen using hollow fibers containing oily rejuvenator.

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## 1. Introduction

Some irreversible aging and damage can be found because of the influence of environmental factors or stress in the course of using materials [1]. These injuries are often hidden in the material and are difficult to come to light. Long term durability and reliability are critical to the materials used in the structure [2]. Self-healing function is one of the most basic characteristics of nature and self-healing materials are still considered to be an important area of research in next years [3,4]. The self-healing researches are carried out in nearly all types of materials, such as ceramics, metals, polymers, and cementitious materials. Self-healing can be divided into two classes according to the healing mechanism, namely intrinsic and extrinsic self-healing materials [5]. The intrinsic ones can be obtained through different chemical reaction approaches, such as photo inducement, molecular inter diffusion reversible bond formation, and recombination of chain ends [6]. On the contrary, the extrinsic ones may do not possess an intrinsic self-healing capability or owe a relatively weak self-healing capability. In other words, these materials need external healing components to achieve the self-healing function by deliberately embedding microcapsules and hollow fibers containing healing agents [7].

In recent years, the concept of self-healing has been extended to the engineering and construction fields [8]. Interestingly, it is worth mentioning that the self-healing bitumen has attracted increasing attention in self-healing materials because this smart self-healing technology is considered as a potential revolution for the pavements. Predictably, self-healing materials may convert the construction and maintenance of pavements through increasing the service time and decreasing the maintenance cost [9]. Self-healing bitumen can reduce the dosage of resources in pavement maintenance, decrease the traffic blockage during maintenance, decrease pollutant and greenhouse gases emissions and elevate road safety and lifespan [10]. Undoubtedly, the self-healing technology will enhance the level of intelligence of roads for the future. At present, the bituminous pavement designs follow the principles of enhancing performance, increasing durability and improving load-carrying capability. With the progress of intelligent science and technology, the bitumen pavement design may be motivated efforts to accomplish a goal allowing roads repair themselves to a certain extent of original state [11].

In general, the self-healing capability of bituminous pavements can be improved by external additives. To achieve the above goal, these additives must survive and prevail in the harsh conditions during treatment process and service life in asphalt [12]. To date, a literature review shows that two methods are considered as the effective approaches for the self-healing bitumen. The first one is called as heat induction method, which has gained popularity in self-healing bituminous material research [13]. Electrical conductive fibers and fillers were added into bituminous pavement and heat produced by electric current enhancing the self-healing ability of bituminous materials. Another method is the usage of microcapsules containing oily rejuvenator [14] or hollow fibers containing rejuvenator [15]. The self-healing mechanism of bitumen applying microcapsules containing rejuvenator has been mentioned systemically in our previous works [11,14,16]. Several microcapsules could be punctured by the tip-stress of a microcrack in its propagation path. The encapsulated oily rejuvenator then was released and moved into the gap of the cracks through capillary. Small molecules of rejuvenator then diffused into the aged molecules of bitumen and softened the bituminous binders. The viscous flow facilitated a healing process and prevented a further propagation of the microcrack [17]. The self-healing processes mentioned above were also repeatable as a multi-self-healing,

which had been proved in previous works [14]. In particular, the microencapsulated waste cooking oil was recycle-used as self-healing additive in asphalt. This idea has potential environmental value and technological value. Besides the capsules system, hollow fibers containing rejuvenator also have been fabricated for self-healing bitumen. Tabaković [15] fabricated sodium alginate fibers within bituminous rejuvenator for self healing bitumen by electrospinning method. Results showed that these compartmented fibers demonstrated good thermal and mechanical strength. Limited by the spinning technology, the hollow fibers did not have a continuous hollow cavity. When rejuvenator is encapsulated in fibers, this structure may block the smooth flow of liquid rejuvenator. Sodium alginate has a dehydration process at 60–170 °C. Therefore, it can be deduced that these sodium alginate fibers may not resist the high temperature (180 °C) of bitumen during pavements. In addition, the fibers prepared by electrospinning are difficult to ensure the strength of the fibers satisfying the application in bituminous pavement. As a biomaterial extracted widely from the cell walls of brown algae, the sodium alginate also will biodegrade in nature losing its original shape and material. To overcome the above shortcoming, Su [18] reported a preparation method of continuous hollow fibers containing rejuvenator by a spinning technology using polyvinylidene fluoride resin (PVDF). Because of the existence of fluorine element, the fibers had an outstanding thermal stability and mechanical property.

Both approaches of using microcapsules or hollow fibers are based on the supplementation of small molecules into aged bitumen. Bituminous material unavoidably loses part of visco-elastic character after a few years. The molecules of rejuvenator will soften the aged bitumen and recovery the visco-elastic capability. Self-healing phenomena normally appear in the binders of asphalt mixture. However, no released rejuvenator molecules can penetrate into aged materials without breaks of microcapsules or hollow fibers. In other words, the self-healing will not happen when no crack has appeared. The original properties of bitumen are lost during a long service time suffering external environmental factors. Predictably but disappointingly, the mentioned microcapsules or hollow fibers methods are only remedial measures when cracks have already generated. Therefore, it is perfect approach to take preventive measures in advance. We know that human skin will be aging caused by many internal and external causes and it becomes thinner, less elasticity and more easily damaged. An effective anti-aging method is using skin creams, which are designed to supply small molecules for skins reducing or diminishing the effects of aging. Inspired by the skin nutrition concept, it will be an ideal method that the rejuvenator can be timely supplied when the bitumen are aging. For the first time, this self-nourishing concept is mentioned in this work to describe this biomimetic behavior. It means that the rejuvenator can be controlled releasing into the aged bitumen. The microcrack can be avoided to be triggered at the greatest extent possible as the bituminous material keeps a softened state. The hollow fiber-based self-healing method is better for polymeric materials. Similar to the vasculs in human body, the hollow fibers forming networks in a material also can be used to supply healing agent to damages [15]. The crack propagation will definitely pass through the networks and trigger breaks of fibers. Then the contained liquid in hollow fibers flows out and heals the cracks. This process owns a corresponding mechanism of microencapsulated rejuvenator. However, vascular systems obviously have more advantages over microencapsulated rejuvenator based systems. The vascular systems can ensured the release of rejuvenator into cracks, because the crack has a higher probability to pass through fiber network. Moreover, the vascular systems can supply large volumes of liquid agents continuously as it has a continuous pipe structure. In addition, more liquid may potentially

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