Evaluation of sunflower oil's healing effect in bituminous materials

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Abstract. Recently, sunflower oil has been widely used as encapsulated healing agent for self-healing asphalt. However, few researches have evaluated the healing effect of sunflower oil on aged asphalt binder. As such, this paper investigates the potential healing effect of sunflower oil in bituminous materials. Dynamic shear rheometer (DSR) was employed to evaluate the performance of the asphalt binders. The resultant master curve indicated that by adding 5% sunflower oil, approximately 30% loss of modulus in aged bitumen could be restored. Thus, the test result bear solid testimony that sunflower oil has the potential to fully recover the original rheological properties as well as healing capacity of the asphalt binder.

Keywords: Sunflower oil, Self-healing asphalt, Dynamic shear rheometer, Rejuvenator, Asphalt behaviour.

1. Introduction
Due to its special visco-elastic property, asphalt concrete has been widely used as the paving material throughout the world. In its service life, asphalt pavement suffers damage from cyclic loadings and it ages due to environmental factors such as oxidation, moisture and long exposure to UV light. As a result, asphalt pavements will become stiffer and more brittle with the increasing time into its service life. Though asphalt pavement has been proven to have the ability to heal itself, the long rest period needed for asphalt to do so often made this option insufficient. However, studies undertaken to address this insufficiency have shown that the self-healing process of asphalt pavements can be accelerated by addition of asphalt rejuvenator [1-4]. This is because ageing will increase the stiffness of asphalt pavements and the addition of rejuvenator will reverse this process. In industry, rejuvenator can be sprayed on the surface layer of asphalt pavements to refresh it thus prolong its service life, but it only offers top layer treatment as rejuvenators are unable to penetrate more than 20mm of the surface layer [5]. To solve this problem, the capsule healing systems were developed and introduced in asphalt pavement. In these systems, rejuvenators are encapsulated in compartmented fibres or capsules which are then embedded evenly throughout asphalt pavements [6-7]. When micro-cracks in the pavements propagate through the capsules, the tension created will rupture the capsule and trigger the release of rejuvenator [8]. The visco-elastic behaviour of the asphalt will be improved to allow the asphalt flow more easily, thereby closing the cracks. Generally, typical healing agents used for asphalt rejuvenation include industrial rejuvenator, waste cooking oil and vegetable oil.
Aimed to rejuvenate the aged asphalt binder and recover its original properties, the industrial rejuvenators are widely used in the recycling of asphalt concrete and surface layer regeneration treatments. A. Tabakovic et al. [3] used industrial rejuvenator as an encapsulated healing agent in compartmented fibre recently. In the paper, the authors demonstrated that compartmented fibres can release controlled amount of rejuvenator that result in multiple effective healing. With industrial rejuvenator, the healing efficiency of the tested asphalt mixture was improved by 40%.

Waste cooking oil (WCO) is a type of non-edible oil that is often regarded as food waste. However, the physical and chemical properties of WCO are almost similar to fresh cooking oil [9]. Therefore, WCOs has a wide range of real-life applications including bio-fuels and soap [10]. Due to the availability and accessibility of WCO, a research regarding the use of WCO as rejuvenator for asphalt mixtures was conducted by Asli et al. [11]. Using penetration value, softening value and viscosity, the authors demonstrated that waste cooking oil can achieve rejuvenation in various types of asphalt to restore their original characteristics. Furthermore, waste cooking oil can reduce the asphaltenes to the maltenes ratio but not till an extent that it resembles that of virgin bitumen.

Vegetable oils can rejuvenate a wide variety of asphalts because they are compatible with penetration grade bitumen. Vegetable oils are also environmentally friendly when mass produced thus ideal to be used in large quantity. Among all the vegetable oil that has been added as the encapsulated healing agent in studies, sunflower oil is most widely used. Garcia et al. [12] demonstrated that addition of capsules containing sunflower oil into asphalt mortar result in effective self-healing and improvement in its healing capacity. Another successful application of sunflower oil as the encapsulated rejuvenator was done by Shizad et al. [13-14]. The authors showed that encapsulating sunflower oil in double-walled PU/UF microcapsules was effective in decreasing stiffness of asphalt binders thus asphalt mortars. Furthermore, asphalt mixture at room temperature showed higher healing efficiency at day 1 but the difference slowly tapered off as time passed by. By using a three-point-bending testing and healing programme, a study by T. al-Mansoori et al. [15] demonstrated that healing level of asphalt beam is higher with addition of microcapsules encapsulating sunflower oil and is positively correlated to the number of microcapsules introduced in the beams, which indicates that the healing level can be controlled by the applied amount of sunflower oil.

Although sunflower oil has been wildly used for asphalt self-healing, few researches have quantified its healing effect on aged asphalt binder. Hence, this paper focuses on investigating the healing effect of sunflower oil on aged bitumen sample through testing its rheology properties using Dynamic shear rheometer (DSR).

2. Experimental methods

The performance of asphalt pavement was evaluated using a dynamic shear rheometer (DSR) in this paper. Virgin bitumen was first tested using DSR to obtain its complex shear modulus ($G^*$). Then, the asphalt ageing phenomenon were simulated using rolling thin-film oven (RTFO) and pressure ageing vessel (PAV). The aged bitumen was again tested with DSR to evaluate its ageing level. Lastly, various contents of sunflower oil were added to the aged bitumen samples and the rejuvenated bitumen samples were tested with DSR to investigate their healing effect. The whole process can be summarised in Figure 1 below.
Materials
The asphalt binder used in this study was made of standard 70/100 bitumen provided by Q8, a Kuwait Petroleum Corporation. Commercially available sunflower oil was purchased from the supermarket and used in this study.

Ageing methods
RTFO
The Rolling Thin-Film Oven (RTFO) was employed in this study according to AASHTO T 240 to simulate the ageing of asphalt binder during its production process. The test was performed using RTF 325-A from James Cox & Sons Inc. Virgin bitumen was first heated to 160°C and then poured into cylindrical glass bottles which are used in the RTFO. The test was then conducted at 15 rpm for 85 minutes with the oven temperature maintained at 163°C. After cooling, the bitumen sample was scraped from the glass bottle and stored for PAV testing.

PAV
The Pressure Ageing Vessel (PAV) was then employed in this study according to AASHTO R 28 to simulate the ageing of asphalt binder after 7 to 10 years in service. The test was performed using PAV3 from Applied Test Systems Inc. Aged bitumen sample from RTFO test was first heated to 160°C and then poured into a thin oven pan. The pan was then heated in the PAV at 100°C and under a pressure of 2.07 MPa for 20 hours which marks the end of the ageing process.

Addition of sunflower oil
When mixing the aged bitumen with sunflower oil, the aged bitumen was first heated to 160°C and then sunflower oil was added to the samples at a range of 1% to 5% by weight, respectively. Thereafter, the mixtures were stirred at 300 rpm for 5 minutes and the rejuvenated bitumen samples were prepared.

DSR

Figure 1. Experimental Process
In this study, virgin bitumen, aged bitumen and rejuvenated bitumen were all tested with DSR respectively to compare their performance. The test was performed using Dynamic mechanical analyzer from Anton paar according to AASHTO Provisional Standards, April 2000 Edition. The DSR test was performed under controlled-strain condition at a range of temperatures from 0°C to 40°C using a range of frequency sweep from 0.0628 to 314rad/s (0.01Hz to 50 Hz). The loading plate used in the DSR had a diameter of 8mm and a 2mm gap in between adjacent parallel plates. Major parameters obtained from DSR were the values of phase angle ($\delta$) and complex shear modulus ($G^*$) from which the rutting parameter and fatigue parameter can be deduced, where:

- Rutting parameter $= \frac{G^*}{\sin \delta}$ and
- Fatigue parameter $= G^* \sin \delta$.

Since asphalt’s visco-elastic behavior satisfy time-temperature superposition, a master curve can thus be constructed to illustrate asphalt performance across a wider frequency at a single temperature.

To construct the abovementioned master curve, the following formulas were used:

1. **Williams–Landel–Ferry equation**

   \[
   \log \alpha_T (T) = \frac{-C_1(T - T_0)}{C_2 + (T - T_0)}
   \]

   Where:
   - $\alpha_T =$ superposition parameter,
   - $T =$temperature,
   - $T_0 =$reference temperature chosen to construct the compliance master curve,
   - $C_1, C_2 =$empirical constants.

2. **Symmetrical Sigmoidal Equation**

   \[
   \log |G^*|_{fit} (f_r) = \log |G^*|_{min} + \frac{\log |G^*|_{max} - \log |G^*|_{min}}{1 + e^{\beta + \gamma (\log f_r + \log \alpha_T)}}
   \]

   Where:
   - $G^* =$complex modulus,
   - $f_r =$reduced frequency,
   - $\beta$ and $\gamma =$shifting parameter,
   - $\alpha_T =$superposition parameter,
   - $|G^*|_{max} =$assumed maximum complex modulus,
   - $|G^*|_{min} =$assumed minimum complex modulus,
   - $|G^*|_{fit} =$fitted complex modulus.

3. **Fitting equation**

   \[
   \sum_{n=1}^{N} \text{error} = \sum_{n=1}^{\infty} \left(\frac{\log |G^*|_{test} - \log |G^*|_{fit}}{\log |G^*|_{test}}\right)^2
   \]

   Where:
   - $G^* =$complex modulus,
   - $|G^*|_{test} =$complex modulus obtained from experimental result.
   - $|G^*|_{fit} =$fitted complex modulus,
   - Error = Fitting error.

### 3. Results and Discussions

All experimental data in this study are solely collected from the DSR to characterise the behaviour of bitumen samples. By fitting the data acquired into a master curve, the ageing and healing level of the bitumen samples can be quantified. Also, the rutting parameter and fatigue parameter of the bitumen samples could be compared. The rutting parameter could characterise asphalt rutting resistance under high temperature while the fatigue parameter could characterise asphalt performance against cyclic loadings.
Ageing of virgin bitumen

The master curve shown in figure 2 was constructed by scaling the modulus curve using formulas introduced in section 2.4. Figure 2 indicated that the complex modulus of all bitumen samples increases as the loading frequency increase. However, the curve of aged bitumen sample is always above the curve of virgin bitumen sample. This indicates that the complex modulus of aged binder is significantly higher than that of virgin bitumen under all testing frequencies. It can thus be concluded that compared to the virgin bitumen, the aged bitumen is stiffer as more stress is required to produce the same level of strain in aged bitumen. This shows the standard ageing procedure used in this paper successfully simulated the ageing process of virgin bitumen in the field.

Figure 2. Master curve of the virgin bitumen and aged bitumen samples

Healing effect of sunflower oil

In Figure 3, the complex modulus increases with increasing loading frequency for all curves with or without sunflower oil. However, the addition of sunflower is shown to be effective in reversing the increment of complex modulus caused by the ageing procedure. Under low frequency area, the aged bitumen sample is able to recover 13.69% of its original modulus with 1% addition of sunflower oil. Further increasing the oil content will consistently recover the magnitude of G* until at 5% it resembles that of virgin bitumen with 32.34% of its lost property.

Figure 3. Healing effect of the sunflower oil
Rutting parameter

Table 1 gives the SHRP rutting parameter values for three different loading frequencies at 0.0628, 4.44, 314 rad/s (0.01Hz, 0.71Hz and 50Hz). The data tested at 20°C were selected to represent the normal serving temperature of asphalt pavement. Five different types of samples used in the tests include: 70/100 virgin bitumen, aged bitumen and another three bitumen samples rejuvenated with 1%, 3% and 5% sunflower oil. Table 1 demonstrated that the value of the rutting parameter increases as bitumen ages but the addition of sunflower oil will reduce that value. Additionally, all the rejuvenated bitumen samples have a lower frequency sensitivity than the aged bitumen sample.

Although addition of sunflower oil will decrease the rutting parameter of the sample bitumen, asphalt behaviour is also temperature dependent and a lower rutting parameter will improve asphalt resistance against fracture at low temperature.

| Type            | 0.0628 rad/s | 4.44 rad/s | 314 rad/s |
|-----------------|--------------|------------|-----------|
| Virgin Bitumen  | 4.64E+04     | 1.43E+06   | 2.58E+07  |
| Aged Bitumen    | 4.32E+09     | 6.21E+10   | 5.85E+11  |
| 1% Sunflower Oil| 2.83E+05     | 4.39E+06   | 4.04E+07  |
| 3% Sunflower Oil| 2.37E+05     | 3.81E+06   | 3.69E+07  |
| 5% Sunflower Oil| 1.79E+05     | 3.01E+06   | 3.17E+07  |

Fatigue parameter

Table 2 provided the SHRP fatigue parameter values of all five previously mentioned groups of bitumen samples under 20°C with three loading frequencies at 0.0628, 4.44, 314 rad/s (0.01Hz, 0.71Hz and 50Hz). Table 2 indicated that the value of the fatigue parameter increases as bitumen ages but the addition of sunflower oil will reduce that value. Similar with section 3.3, all the rejuvenated bitumen samples have a lower frequency sensitivity than the aged bitumen sample.

The test result thus quantified for us the extent to which the addition of sunflower oil can improve the fatigue resistance of the aged bitumen.

| Type            | 0.0628 rad/s | 4.44 rad/s | 314 rad/s |
|-----------------|--------------|------------|-----------|
| Virgin Bitumen  | 4.49E+04     | 1.22E+06   | 1.42E+07  |
| Aged Bitumen    | 3.40E+09     | 3.68E+10   | 2.14E+11  |
| 1% Sunflower Oil| 2.32E+05     | 2.72E+06   | 1.68E+07  |
| 3% Sunflower Oil| 1.96E+05     | 2.43E+06   | 1.60E+07  |
| 5% Sunflower Oil| 1.50E+05     | 1.99E+06   | 1.40E+07  |

4. Conclusion

This paper investigated the healing effect of sunflower oil on bituminous materials. Rheological properties of asphalt binder were collected to construct a master curve and deduce the rutting/fatigue parameter of the bitumen samples. Conclusions arrived in this paper include:

- The complex modulus of aged bitumen is higher than that of virgin bitumen. This indicates that the complex modulus increases as the bitumen sample ages. Thus, PAV and RTFO procedure can successfully simulate the ageing process of asphalt pavement during its service life.
- The complex modulus of rejuvenated bitumen is lower than that of aged bitumen. This indicates that the addition of sunflower could lower the complex modulus of the aged binder. In conclusion, 32.34% of the rheological properties of aged asphalt could be restored to resemble that of virgin bitumen and thus proved sunflower oil to be an effective healing agent.
The rutting parameter increases as bitumen ages but the addition of 5% sunflower oil will significantly reduce that value to $1.79 \times 10^5$ at 0.0628 rad/s, $3.01 \times 10^6$ at 4.44 rad/s and $3.17 \times 10^7$ at 314 rad/s. Hence, the addition of sunflower oil will improve fracture resistance of bitumen at low temperature.

The fatigue parameter increases as bitumen ages but the addition of 5% sunflower oil will reduce that value to $1.50 \times 10^5$ at 0.0628 rad/s, $1.99 \times 10^6$ at 4.44 rad/s and $1.40 \times 10^7$ at 314 rad/s. The test result thus provided us a quantified prove that the addition of sunflower can improve the fatigue resistance of the aged bitume.

The results of this study quantified the effect of sunflower oil on rheological properties of bituminous material. It also demonstrated the possibility to fully recover the original rheological properties as well as healing capacity of the asphalt binder by adapting the application amount of sunflower oil to the asphalt ageing level. As a result, this study provides evidence and lays solid foundation for studies that use sunflower oil as a healing agent in bituminous material.

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