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Article

The Effect of Ageing on Chemical and Mechanical Properties of Asphalt Mortar

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Abstract: Asphalt mortar is a mixture of bitumen, filler, and sand. Mortar plays an important role in asphalt mixtures as it serves as the adhesive between the coarser aggregates. Due to the effect of bitumen ageing, the chemical and mechanical properties of asphalt mortar evolve with time. The mortar becomes more brittle and prone to cracking, thus leading to inferior pavement performance. In this study, Fourier transform infrared (FTIR) spectrometry was used to quantify changes in the chemical functional groups related to ageing and to calculate the carbonyl and sulfoxide indices. In addition, frequency sweep tests and uniaxial tension tests were performed by means of dynamic shear rheometer (DSR) tests to determine evolution of the stiffness and strength due to ageing. Two different oven ageing protocols were used to evaluate the effect of fine mineral particles on bitumen ageing. The protocols differed with respect to the order of ageing and mixing of the constituents. The results showed that both the chemical and mechanical properties of mortars significantly changed with ageing. Specifically, the carbonyl index, stiffness, and strength of the mortar increased. Under the same ageing conditions, a higher ageing level was observed for mortars produced by first mixing and then ageing compared to the mortars produced by mixing aged bitumen with filler and sand. This could be due to the presence of sand and filler particles, which resulted in an increased length of diffusion paths and consequently a slower ageing process.

Keywords: asphalt mortar; ageing; FTIR; stiffness; strength

1. Introduction

Ageing of bitumen is believed to be a major contributor to many pavement distresses, such as raveling, cracking, and fatigue [1–6]. Ageing occurs during the process of storage, mixing, transportation, construction, and service. Studies have demonstrated that ductility and penetration of bitumen diminish as a result of ageing, while softening point and ignition temperature increase in the process [7–9]. Moreover, the viscosity increases and bitumen becomes stiffer.

Asphalt mortar, commonly known as fine aggregate matrix, is a mixture of bitumen, filler, and sand. It is the binding medium that holds the aggregates of an asphalt mixture together, and it plays an important role as the link between bitumen properties and mixture performance. Past studies have shown that the testing of bituminous mortar is an efficient and repeatable approach to predict the performance of asphalt mixture [10–12]. The ageing of bitumen, through the asphalt mortar as an intermediate medium, causes the mixture to become excessively hard and brittle and consequently more susceptible to producing pavement distress [13]. For this reason, some studies have considered asphalt mortar ageing. For instance, Cui et al. investigated the effect of laboratory long-term ageing on the rheological properties of asphalt mortar. The long-term ageing test is a combination of

ultraviolet ageing, thermal ageing, and humidity [14]. Molenaar et al. studied the effects of ageing on the mechanical characteristics of bituminous binders in porous asphalt (PA) concrete. The results showed that ageing increased the tensile strength of the bituminous binders but decreased the strain at break [15]. In addition, the fatigue results showed that the adhesive zone between stone and mortar tended to be the weakest link because there was a bitumen-rich interlayer [16]. Tong et al. evaluated the moisture and ageing damage of asphalt mortar using the repeated direct tensional test method. The results showed that fatigue crack growth of asphalt mortar increased substantially after ageing [17]. Zhang et al. quantified the ageing effect on the rheological characteristics of two types of asphalt mortar and applied the results to evaluate the raveling resistance of PA wearing courses in finite element simulations. It was found that ageing had more influence on the complex shear modulus of the base mortar compared to the SBS (Styrene-Butadiene-Styrene) mortar, but its effects on fatigue resistance were the opposite [18]. These studies proved that mortar ageing had a negative influence on pavement performance, such as fatigue and raveling. However, the studies were limited in considering the rheological changes of asphalt mortar subject to various conditions.

The main objective of this study was to investigate the effect of filler and sand on ageing and determine the evolution of mortar properties due to ageing. To achieve this, two different ageing conditions were applied on mortar by an oven. The ageing method differed with respect to the order of ageing and mixing of the constituents. The chemical composition of asphalt mortar was evaluated by means of Fourier transform infrared (FTIR) spectrometer. Cylindrical mortar samples were then prepared and tested using dynamic shear rheometer (DSR) tests. In addition, the original bitumen was tested after the same ageing conditions as that of asphalt mortar.

2. Materials and Methods

2.1. Materials and Asphalt Mortar Mix Design

The PEN 70/100 bitumen, which is one of the most commonly used bitumen in the Netherlands, was used in this study. Table 1 shows the main physical and rheological properties of the PEN 70/100 bitumen. The mortars were prepared using river sand, which is a type of sand with quartz, with a nominal maximum size of 2 mm and Wigro 60K filler, which is a type of filler with hydroxide that contains 25% hydrated lime (density is 2780 kg/cm³).

Table 1. Specifications of PEN 70/100 at fresh (unaged) state.

Property	Unit	PEN 70/100
Penetration at 25 °C	0.1 mm	70–100
Softening point	°C	43–51
Dynamic viscosity at 60 °C	Pa s	160
Complex shear modulus at 1.6 Hz and 60 °C	kPa	1.8
Phase angle at 1.6 Hz and 60 °C	°	88

The mortar was designed based on the mix design of two types of asphalt mixtures—porous asphalt (PA) 16 and stone mastic asphalt (SMA) 16—which represent an open and a dense-graded asphalt mixture, respectively, and are widely used in the Netherlands. Both PA 16 and SMA 16 consist of crushed stones with a nominal maximum aggregate size of 16 mm, river sand, Wigro 60K filler, and PEN 70/100 bitumen. The binder contents were 5.0% and 6.4% for the PA and the SMA mixture, respectively. The aggregate gradations of PA16 and SMA 16 mixtures are shown in Figure 1.

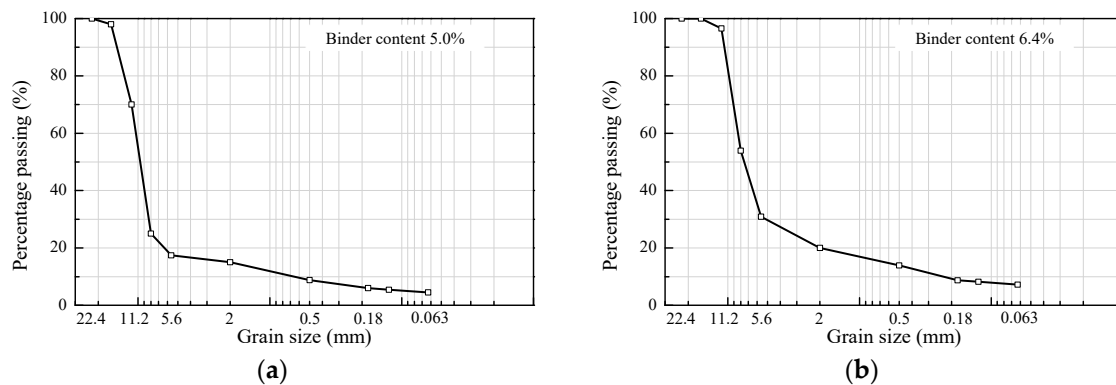


Figure 1. Aggregate gradation and physical properties of mixture: (a) porous asphalt (PA) 16; (b) stone mastic asphalt (SMA) 16.

The asphalt mortar used in this study contained sand with size smaller than 2 mm. On the basis of gradations of PA 16 and SMA 16 in Figure 1, the mass ratios of bitumen, filler, and sand of each mixture were calculated and are presented in Table 2. Specifically, it was 1:0.9:2.1 for the mortar in PA 16 and 1:1.25:2 for the mortar in SMA 16. Therefore, considering an average between the mortars in PA 16 and SMA 16, the mass ratio of each composition in the designed mortar was selected as 1:1:2. This mortar composition was employed in the lab investigation.

Table 2. Asphalt mortar recipe.

Mix Design	Bitumen	Filler	Sand
Mortar in PA 16	1	0.9	2.1
Mortar in SMA 16	1	1.125	2
Designed Mortar	1	1	2

2.2. Ageing Program

First, the mortar samples were tested without applying any ageing protocol so as to define their properties at unaged (fresh) conditions. Then, two oven ageing protocols that were different in the order of ageing and mixing of the constituents were applied. Specifically, in the first method, the mortar constituents—bitumen, filler, and sand—were mixed after they were aged separately in the oven (denoted as AM); this method is commonly used as laboratory ageing protocol. In contrast, the second method involved first mixing the constituents to prepare the mortar and then subjecting the mortar samples to ageing (denoted as MA); this method is close to the ageing condition in the field. The purpose of having two different protocols was to identify the effect of filler and sand on ageing. Table 3 summarizes the ageing protocols that were considered. Specifically, the temperature was selected as 100 °C for all ageing tests, and the time varied from 20 to 160 h. In addition, bitumen samples after the application of various ageing times were studied.

Table 3. Ageing program.

Ageing Temperature (°C)	Ageing Time (h)	Mortar_AM	Mortar_MA	Bitumen
100	20	✓		✓
	40	✓	✓	✓
	80	✓		✓
	160	✓	✓	✓

2.3. Sample Preparation

As the studied mortar samples were composed using fractions of aggregates up to 2 mm, the parallel-plates configuration could no longer be applied. Instead of the parallel-plates configuration,

a special set-up with clamps for cylindrical mortar sample was used for the DSR tests [19,20] (Figure 2a). The preparation procedure of the mortar samples was adopted from a previous study at TU Delft [21]. Initially, the PEN 70/100 bitumen, filler, and sand were separately heated to 150 °C. Then, the three compounds were mixed manually for 10–15 min. After the mixing process, asphalt mortar was poured into a silicone mold (Figure 2b). Two metal rings were prefixed on the top and bottom side of the mold. These metal rings ensured load transfer from the DSR into the samples (Figure 2c). After filling the mold with mortar, the mold was placed in the oven at 150 °C for 5 min to allow the suppression of possible air pockets that were formulated during pouring of the mix into the mold. Afterwards, the mold was cooled down to ambient temperature and then placed in a refrigerator at −15 °C for at least 2 h. Finally, the mortar samples were removed from the mold and checked visually for air voids at the surface of the sample. The accepted samples were placed in a special silicone mold and stored in the refrigerator at −15 °C before testing (Figure 2d).

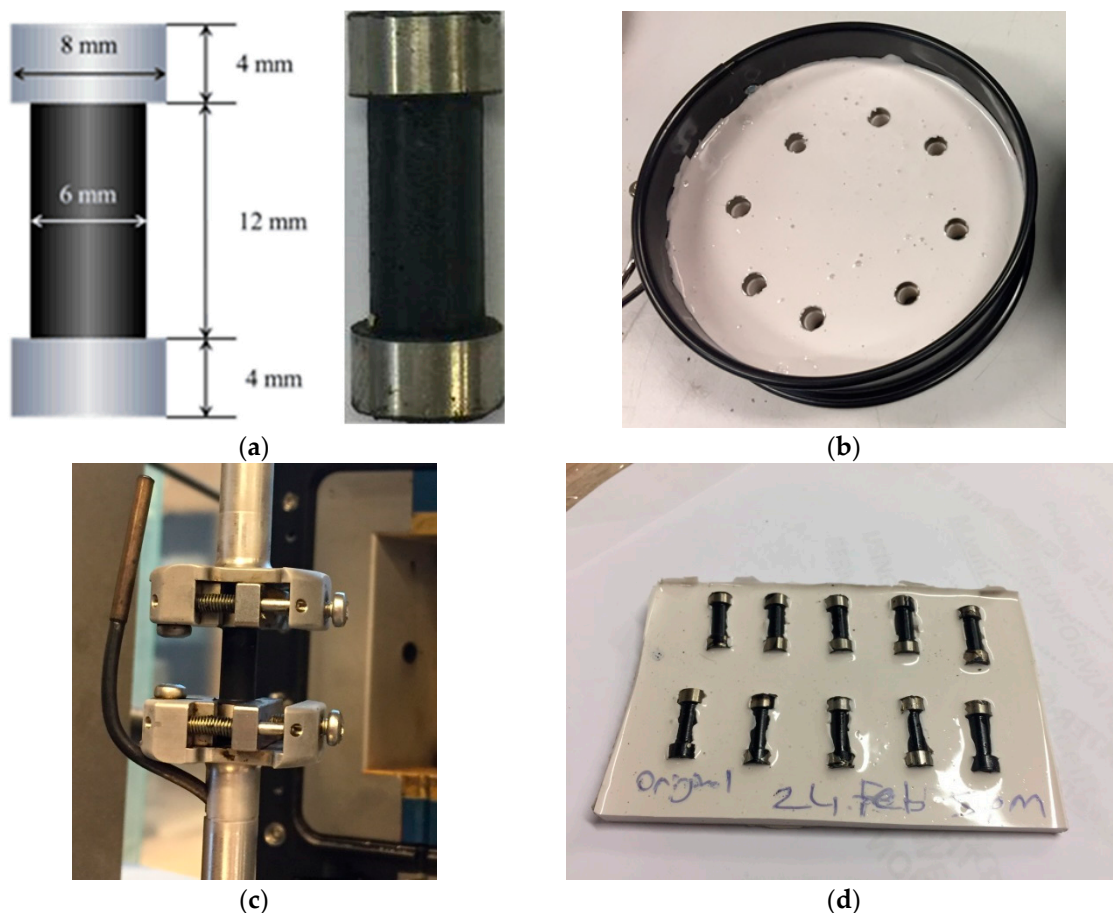


Figure 2. Asphalt mortar sample: (a) geometry of sample; (b) silicone mold for sample preparation; (c) measuring system for cylinder sample; (d) silicone mold for sample storage.

2.4. Experimental Methods

As an efficient, convenient, and inexpensive tool, the FTIR is widely used to determine the chemical characteristics of bituminous materials. In the infrared spectrogram of bitumen, the absorbance bands corresponding to carbonyls and sulfoxides show a significant increase with ageing [22–24]. The chemical characterization in this study was performed using the Spectrum 100 FT-IR spectrometer from PerkinElmer. A single-beam configuration was used. The sample was scanned 20 times with a fixed instrument resolution of 4 cm^{−1}. The wavenumbers were set to vary from 600 to 4000 cm^{−1}. At least three repetition tests were performed for each ageing condition.

DSR is commonly used to investigate the rheological properties of bituminous materials [25–27]. In this study, the linear viscoelastic (LVE) strain range of the bitumen and asphalt mortar samples were initially determined using amplitude sweep tests. Within the LVE strain range, frequency sweep tests were performed at four different temperatures (0, 10, 20, and 30 °C). During the tests, the frequency varied in a logarithmic manner from 50 Hz to 0.01 Hz. Uniaxial tensile tests were performed at 0 °C with 0.2% (0.024 mm/s) tensile rate. The load and deformation in the testing process were automatically collected with a frequency of two measurements per second.

3. Results and Discussion

3.1. Fourier Transform Infrared Spectrometer

3.1.1. FTIR Spectra

The FTIR spectra were obtained in a wavenumber range from 4000 to 600 cm^{-1} . However, the changes caused by ageing could be found at wavenumbers lower than 2000 cm^{-1} , as shown in Figure 3. These wavenumbers corresponded to functional groups related to the oxidation processes. It showed that the carbonyl (left, red part) and sulfoxide functional (right, blue part) groups changed considerably after laboratory ageing, i.e., they both increased with ageing.

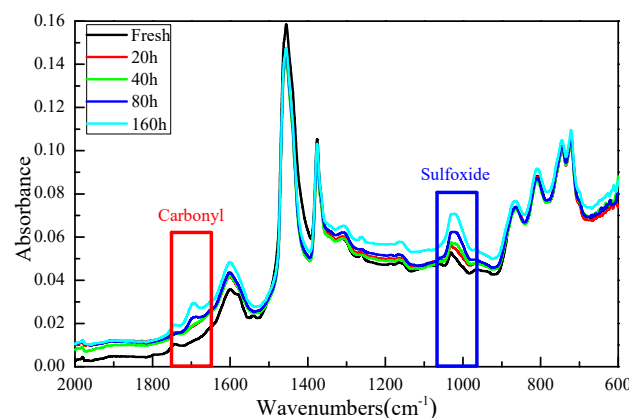


Figure 3. FTIR spectra of PEN 70/100 at different ageing hours.

The FTIR spectra of fresh mortar sample and each component (fresh bitumen, filler, and sand) are shown in Figure 4. It can be observed that the mortar—a mixture of bitumen, filler, and sand—had all functional groups as its three component materials. Figure 4 clearly shows that the carbonyl group that was present in the mortar originated only from the presence of bitumen, whereas sulfoxides can be present in all three components.

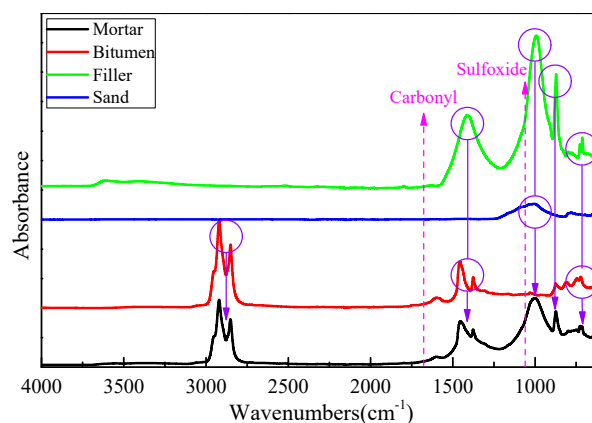


Figure 4. FTIR spectrum of mortar, bitumen, filler, and sand at fresh state.

3.1.2. Ageing Index

In this study, the effects of ageing were analyzed considering specific bands of wavenumber as defined by Lamontagne et al. [28–30] and the corresponding area under those bands. Then, the carbonyl and the sulfoxide indices were determined by dividing the area under a specific location of the spectrum by the sum of specific areas, Equations (1) and (2).

$$\text{Carbonyl index} = \frac{A_{1700}}{\sum A} \quad (1)$$

$$\text{Sulfoxide index} = \frac{A_{1030}}{\sum A} \quad (2)$$

where $\sum A = A_{(2953,2862)} + A_{1700} + A_{1600} + A_{1460} + A_{1376} + A_{1030} + A_{864} + A_{814} + A_{743} + A_{724}$.

Using the obtained spectra, the ageing indices (carbonyl and sulfoxide index) of each sample were calculated. Figure 5 shows the carbonyl and sulfoxide indices (average value of three measurements) of bitumen, mortar_AM, and mortar_MA. The error bar in Figure 5 is the standard deviation of the three measurements. It can be observed that under the same ageing conditions, the value of the carbonyl index for mortar_AM was similar to that of bitumen. This can be attributed to the fact that carbonyls in the mortar only came from bitumen as illustrated in Figure 4. Previous studies have shown that carbonyls are formed later than sulfoxides because sulfur is more reactive than carbon in bitumen. Later, carbonyls increase due to the full consumption of sulfur [31]. Similar trends and values were found for the mortar in our study as well.

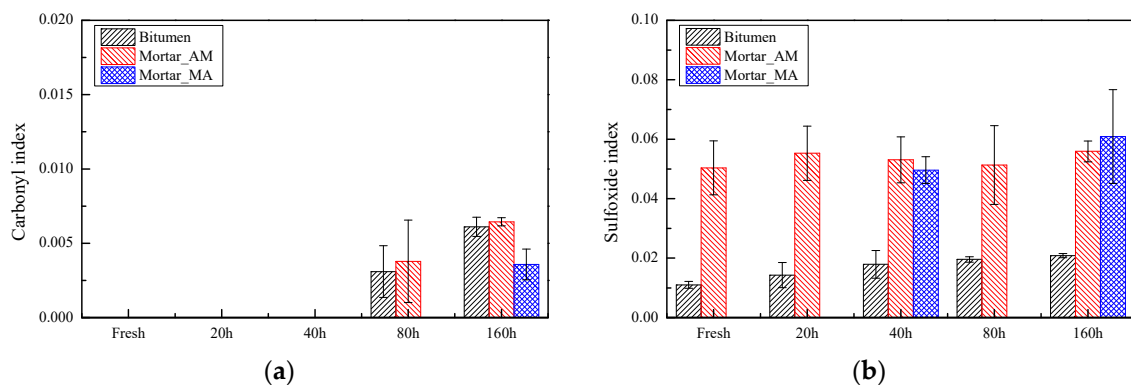


Figure 5. Function group index of mortar at different ageing conditions: (a) carbonyl index; (b) sulfoxide index.

Moreover, it can be observed that the carbonyl index of mortar_MA was significantly smaller than that of mortar_AM (Figure 5a), indicating that mortar_MA had a lower ageing rate. This finding confirms the theory that the sand and filler particles present in the mortar can slow down the diffusion rate of oxygen due to an increased length of diffusion paths and consequently slow down the ageing process as well [32]. In addition, the diffusivity of mortar will decrease as the bitumen volume fraction decreases [33]. This happens because mortar is the mixtures of bitumen, filler, and sand, and its diffusivity should always be between that of the bitumen and the filler/sand. Moreover, the diffusivity of bitumen is much higher than that of filler/sand [34,35].

The sulfoxide index of mortar_AM differed significantly with that of bitumen (Figure 4b). This was because sulfoxides in the mortar came not only from the bitumen component but also from the filler and sand particles as illustrated in Figure 3. The sulfoxides content in filler and sand was much higher than that in bitumen, and this led to obvious change in the sulfoxide index of the mortar. Moreover, as another result, the deviation of the mortar results was obviously higher than that of bitumen. The reason for this might be because the measuring area of the FTIR test was limited, meaning the

infrared radiation hit only one or few of the components of mortar. Another reason might be that the mix of mortar samples was not homogenous.

3.2. Dynamic Shear Rheometer

3.2.1. Frequency Sweep Tests

At least three replicate samples were tested in DSR at each condition. Based on the time–temperature superposition (TTS) principle, master curves of complex shear modulus and phase angle were generated at reference temperature of 20 °C. Figure 6 shows the evolution of the rheological characteristics of bitumen with increased ageing time. The master curves of mortar_AM and mortar_MA at various ageing times are shown in Figure 7.

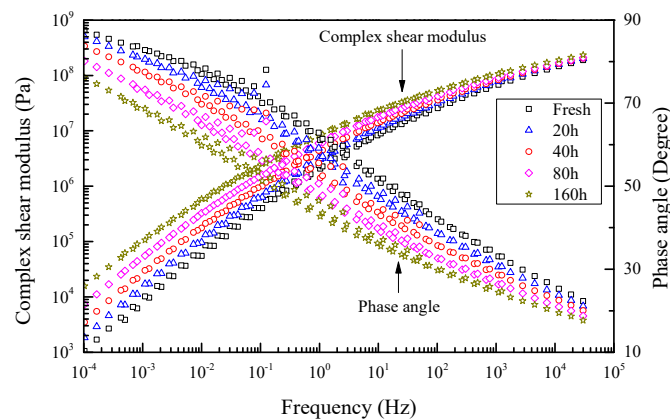


Figure 6. Master curves of PEN 70/100 at different ageing times.

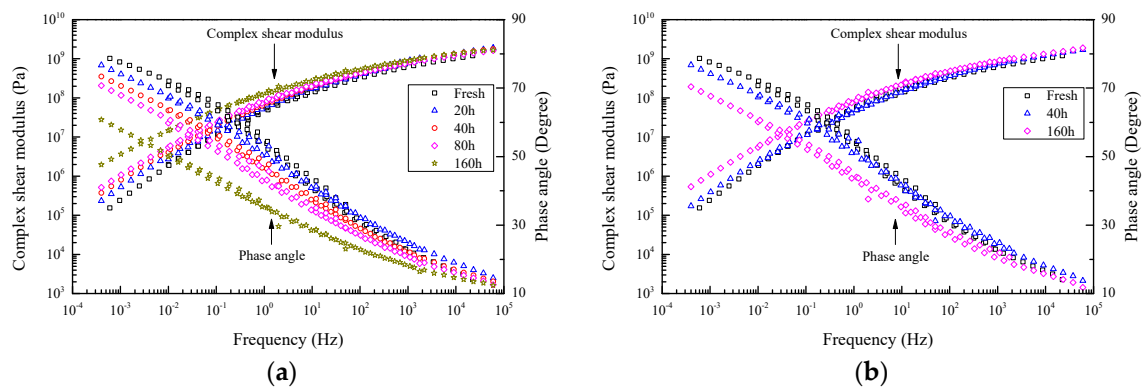


Figure 7. Master curve of mortar samples at different ageing times: (a) mortar_AM; (b) mortar_MA.

As shown in Figure 6, the observed differences were more pronounced at low frequencies, while they were diluted at higher frequencies. Specifically, the variation of complex modulus between fresh and aged materials was about one order of magnitude at low frequencies. Moreover, at the highest frequency, all samples tended to reach an asymptote at a value of 10^8 Pa. On the contrary, the phase angle differed substantially for the whole frequency range. The greater the value of complex shear modulus, the harder was the bitumen; the smaller the phase angle, the more elastic was the bitumen.

In Figure 7, it can be seen that the complex shear modulus of mortar sample increased as expected with increasing ageing, while the opposite was observed for the phase angle. Specifically, the variation of complex modulus between fresh and aged materials was one order of magnitude at low frequencies. Moreover, at the highest frequency, all samples tended to reach an asymptote at a value of 10^9 Pa. The threshold value at high frequency was one order of magnitude larger than that of bitumen due to the addition of filler and sand. On the other hand, the phase angle of the mortar samples differed

substantially for the whole frequency range. These results show that the material became stiffer and more brittle with ageing. The comparison between mortar_AM and mortar_MA is shown in Figure 8.

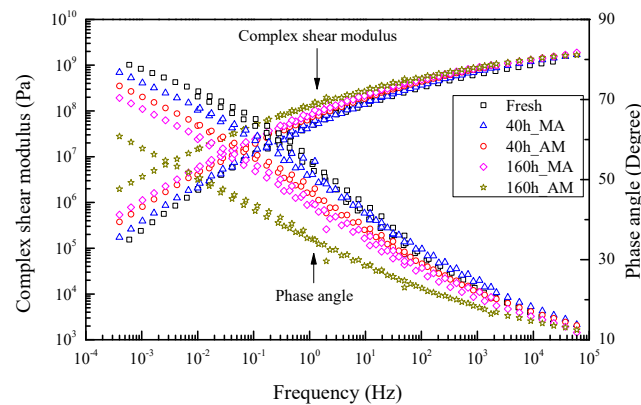


Figure 8. Master curve of mortar_AM and mortar_MA at different ageing times.

Figure 8 shows that compared to mortar_AM, mortar_MA had lower values of complex shear modulus and higher values of phase angle. In other words, mortar_MA was softer under the same ageing conditions, thus the effect of ageing was less. This finding was in agreement with the FTIR results presented in Figure 5. Figure 9 shows the differences in the complex shear modulus and phase angle at 20 °C and at 10 Hz frequency for the bitumen, mortar_MA, and mortar_AM samples. The reason for choosing shear moduli at 10 Hz and 20 °C was that 10 Hz is the commonly used frequency in a dynamic test and 20 °C is considered to be room temperature [36–38].

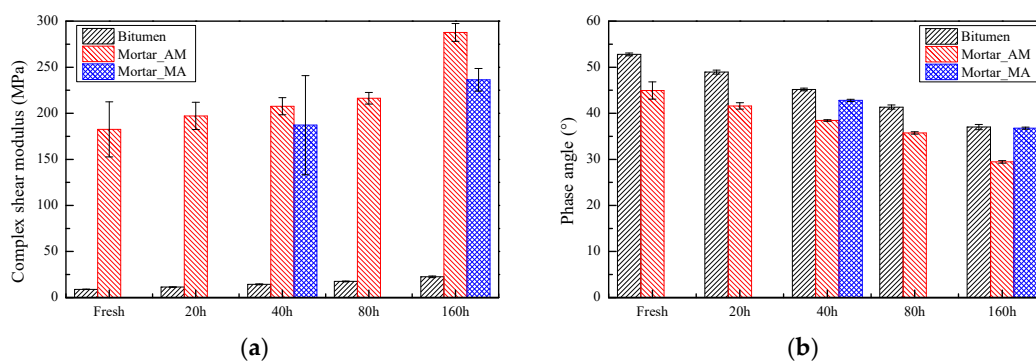


Figure 9. Complex shear modulus and phase angle at 20 °C, 10 Hz of bitumen, mortar_AM, and mortar_MA.: (a) complex shear modulus; (b) phase angle.

It can be observed that the complex shear modulus of mortar was significantly higher than that of bitumen, with the variation being about 200 times higher, while the phase angle of mortar was slightly smaller than that of bitumen. This high difference in stiffness occurred due to the presence of filler and sand particles, while the phase angle of mortar was mainly determined by the bitumen component.

3.2.2. Uniaxial Tension Tests

Uniaxial tension tests were performed using DSR. To obtain the effect of ageing on the tensile behavior of asphalt mortar, at least three samples were tested for each ageing condition. Tensile strength and failure strain are influenced by varying the temperature and strain rate due to the time–temperature dependency of bituminous materials. The tensile strength is higher for lower temperatures and higher strain rates. Therefore, uniaxial tensile tests in this study were performed at 0 °C with 0.2% (0.024 mm/s) tensile rate to make sure all tests can be conducted in a suitable testing range of the device. The normal force in relation to the displacement of the machine for mortar_AM at different ageing times was recorded and is plotted in Figure 10.

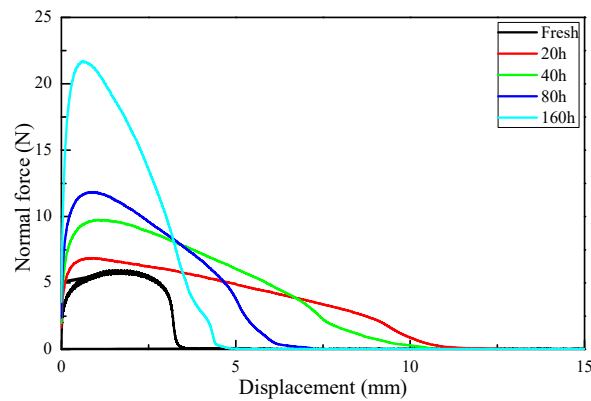


Figure 10. Normal force vs. displacement of mortar_AM with different ageing times.

Figure 10 shows that higher peak normal forces and smaller displacement at failure were obtained for longer ageing times. In other words, aged samples tended to become more brittle. Meanwhile, the steeper initial slopes of the curves can be seen at the longer ageing times, which mean higher moduli for the aged sample. Tensile strength is the maximum normal force divided by the cross-section area of the cylinder sample. It denotes the maximum stress that a material can withstand while being pulled before breaking. Figure 11 shows the tensile strength of asphalt mortar at different ageing times. It can be observed that the strength of mortar increased with ageing time. Another observation was that under the same ageing conditions, the strength of mortar_AM was higher than that of mortar_MA.

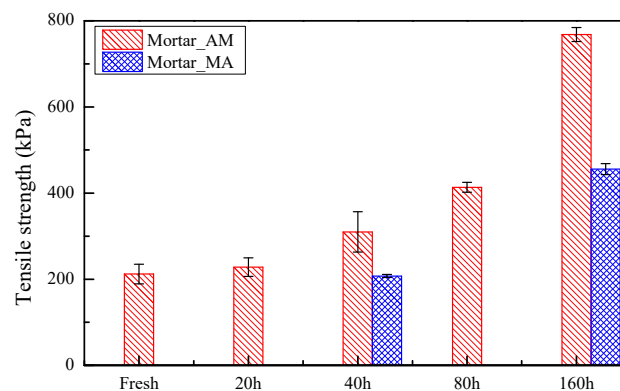


Figure 11. Tensile strength of mortar_AM and mortar_MA with different ageing times.

4. Conclusions

Based on the findings and analysis presented in this paper, the following conclusions can be drawn: (1) Asphalt mortar—a mixture of bitumen, filler, and sand—has all functional groups as its three component materials. Carbonyl group that is present in the mortar originates only from the presence of bitumen, whereas sulfoxides can be present in all three components. (2) Ageing increases the complex shear modulus and decreases the phase angle of bitumen and asphalt mortar. The high difference in stiffness observed between bitumen and asphalt mortar is due to the presence of filler and sand particles. The less difference observed between the phase angle values of bitumen and mortar is due to the fact that the phase angle is mainly governed by the bitumen component. (3) Ageing leads to higher strength and smaller displacement at failure compared to unaged sample. In other words, aged samples tend to become more brittle. (4) Both chemical and mechanical test results have shown that the sand and filler particles present in the mortar may slow down the diffusion rate of oxygen. The hypothesis is that the presence of particles increases the length of the diffusion path and therefore slows down the ageing process. This will help to understand the interaction between components during ageing and develop a more suitable protocol to simulate ageing procedure of bituminous materials in the field.

The results of this study will help to understand the ageing process of asphalt mortar and more accurately predict evolution of its properties due to ageing. Based on the results, the relationship between the chemical and mechanical properties of asphalt mortar can be determined, and a chemo-mechanical model of asphalt mortar can be developed and implemented using the finite element method.

Author Contributions: R.J., X.L., and A.V. conceived the experimental plan and contributed to the interpretation of the results. R.J. carried out the experiments. A.S. and S.E. supervised all project activities.

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Conflicts of Interest: The authors declare no conflicts of interest.

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