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Evaluating the Ageing Sensitivity of the Asphalt Binder via Distinct Ageing Methods

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Abstract: Asphalt binder is a crucial component of asphalt pavements that undergoes ageing over time, which can result in the reduced performance and deterioration of pavements. Consequently, artificial ageing methods play a significant role in providing valuable insights into the ageing behaviour and long-term performance of asphalt binders. However, a consensus on the most effective method for simulating ageing behaviour remains elusive, leading to disparities in the outcomes across different research studies. To address this issue, the study utilises two thermo-oxidative ageing approaches, one focusing on the binder itself and another on the loose asphalt mixture. The study investigates the effect of these ageing methods on the behaviour of asphalt binder using physical, rheological, and chemical characterisation. For the binder ageing method, a rolling thin film oven (RTFO) and a pressure ageing vessel (PAV) were utilised, whereas the loose asphalt mixture ageing was performed in an oven at 95 °C for various durations. The results indicated that the ageing trend differed between the two oxidative ageing approaches as the ageing duration increased. However, by employing an ageing sensitivity index, comparable rheological properties were observed between the binders aged using the PAV for 20 h and the loose asphalt mixture for 5 days. The Fourier Transform Infrared (FTIR) spectroscopy analysis revealed that the ageing methods influenced the functional groups associated with ageing in distinct ways, even though they exhibited similar rheological behaviour. Overall, this study provides a comprehensive understanding of different thermo-oxidative ageing approaches, their correlation, and their relevance to the studied field-aged binders.

Keywords: bitumen ageing; loose asphalt mixture ageing; rheological properties; binder fast characterisation test; long-term ageing; ageing sensitivity index; accelerated ageing

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1. Introduction

Bitumen, often referred to as asphalt binder, is a viscoelastic material derived from the distillation of crude oil. It serves as a binding component in asphalt pavements, holding together other constituents and ensuring their structural integrity [1,2]. However, over time, asphalt pavements undergo oxidative ageing processes due to climatic volatility and chemical reactions. Consequently, the asphalt binder becomes increasingly stiff and brittle, leading to higher susceptibility to cracking and degradation of the pavement [3,4]. It is crucial to identify the ageing mechanism of asphalt binders to implement strategies that can extend the service life of the pavement. In the context of a circular economy, where resource efficiency and sustainability are of utmost importance, investigating the ageing mechanism becomes indispensable for adopting methodologies that enhance pavement performance, optimise resource utilisation, and minimise waste generation [5,6]. Several studies have been conducted to understand the factors that influence the ageing of asphalt pavements and their impact on pavement performance. It has been observed that the effects of ageing have a significant impact on the long-term performance of pavements, which is crucial in the field of road engineering to provide durable and comfortable road surfaces for users [7–9].

In general, the ageing of asphalt mixtures can be categorised into two stages: short-term ageing (STA) and long-term ageing (LTA). The impact of high processing temperatures during the production and construction phases of asphalt mixtures results in the volatilisation of bitumen, known as short-term ageing. The evaporation of volatiles such as saturated and aromatic components is directly proportional to the production temperature of the asphalt mixture. Some previous studies reported that a rise in the production temperature increases the amount of loss of volatility, which has a linear influence on the imbalance of saturate, aromatic, resin, and asphaltene (SARA) fractions. Hence, it increases the stiffness, hardness, viscosity, and fragile nature of the bitumen [10,11]. Simultaneously, long-term ageing occurs over the service life of the pavement due to climatic fluctuations, ultraviolet (UV) radiation, oxygen, and vehicle-induced loads. In this regard, oxidation is one of the key factors influencing the change in the chemical properties of bitumen as it gets exposed to oxygen. The oxidation influences the polar components in bitumen, such as causing asphaltenes to increase, and consequently, they stiffen the binder more, whereas nonpolar components like aromatics that control the flexibility of the binder get reduced due to ageing. Eventually, such ageing stiffens asphalt mixtures and leads to pavement distress [12,13]. The oxidation rate generally depends on factors such as the bitumen content, access to oxygen by the pavement layer, void content, temperature, thickness of the layer, and the existence of cracks [14]. Based on this understanding, researchers have made numerous attempts to simulate field ageing phenomena in the laboratory using various accelerated ageing techniques to evaluate and anticipate the long-term performance of the pavement. Most studies have focused on analysing the influence of temperature, oxygen pressure, or a combination of both parameters to correlate with field performance. Among the ageing techniques, the most widely used laboratory protocol is the asphalt binder ageing, which utilises a rolling thin film oven (RTFO) and a pressure ageing vessel (PAV) to simulate the short-term and long-term ageing mechanisms of asphalt binders, respectively [15,16]. These techniques are often employed in the Superpave performance grading system and serve as valuable tools for comparing and establishing benchmarks for various binders. However, this simulation approach fails to account for the influence of mineral aggregates, which can significantly impact the ageing simulation of asphalt pavement [17]. Although the use of high temperatures and pressure allows for the quick oxidation of bitumen, it cannot effectively replicate field conditions. To address the effects of mineral aggregates in the ageing mechanism, the American Association of State Highway and Transportation Officials (AASHTO) developed a standard asphalt mixture ageing protocol. This protocol can simulate long-term ageing effects in a compacted specimen with a diameter of 100 mm in an oven at 85 °C for 120 h. According to AASHTO R30, this accelerated ageing method simulates a long-term ageing level corresponding to five to ten years of service life. The significance of this method lies in its ability to simulate LTA effects within a reasonable time frame using standard laboratory equipment, making it cost-effective and easier compared to other approaches. However, this method has been criticised for the non-uniformity of ageing throughout the specimen, geometry distortion, and its inability to simulate ageing under various climatic conditions [18,19]. Another advanced ageing technique is the Viennese Ageing Procedure (VAPro), which was developed in Austria. VAPro is considered an extension of the ageing procedure outlined in the SHRP-A-383 report. It utilises a unique configuration consisting of a triaxial cell with a forced flow of highly oxidative gaseous agents such as ozone and nitric oxide over the specimen, at a rate of 1 litre per minute. This approach reportedly simulates a long-term ageing condition closely resembling the field within an efficient timeframe of 4 days [20]. However, this ageing approach utilises an intricate setup for the ageing process, demanding costly configurations.

Furthermore, to address the limitations of previous ageing protocols on compacted specimens, researchers have made efforts to implement accelerated ageing methods for loose asphalt mixtures. The aim is to replicate more realistic field ageing mechanisms and better simulate the characteristics of reclaimed asphalt pavement (RAP). As mentioned earlier, the oxidative ageing mechanism of asphalt binder depends on various contributing factors, particularly temperature and ultraviolet (UV) radiation [21,22]. Several studies have attempted to incorporate UV light into ageing techniques to investigate its influence on the behaviour of bitumen. They have identified that the energy from UV radiation can break down molecular bonds, leading to the surface ageing of the binder [23,24]. Acknowledging the significance of temperature, it is a crucial factor considered in the laboratory simulation of long-term ageing characteristics of asphalt mixtures. The rate of oxidation increases with temperature, and particularly at high temperatures, the ageing may not accurately reflect real-field-aged asphalt binder [25]. A study conducted by the National Cooperative Highway Research Program (NCHRP) reported that long-term ageing can be simulated on a loose asphalt mixture at 95 °C inside a forced-draft oven. The duration of the conditioning period is calculated based on the climatic ageing index (CAI), which depends on the pavement temperature histories at different locations and depths. Data from the Enhanced Integrated Climatic Model (EICM) were used to predict pavement temperature by considering its relationship with pavement depth [19]. However, this approach, while considering pavement temperature histories at distinct locations, still neglects significant dynamic factors such as moisture and traffic conditions during the ageing simulation. This limitation could impact its applicability and its ability to accurately reflect real-field ageing phenomena, which remains a topic of ongoing discussion among researchers.

The literature reveals that several accelerated ageing techniques have been implemented to study the ageing mechanism and long-term performance of asphalt binders. However, there is still no consensus on the most effective approach for simulating ageing behaviour, leading to discrepancies in outcomes among research studies and making it difficult to compare and generalise the findings. In view of these challenges, this study aims to investigate and compare the ageing behaviour of asphalt binder using two different thermo-oxidative ageing approaches, one focusing on the binder itself and the other on the loose asphalt mixtures, with varying ageing durations. Although UV radiation is also a triggering factor for ageing, thermo-oxidative ageing was considered a vital cause, and that accounts for more than 80% of the total life cycle ageing of the bitumen [26]. The ageing sensitivity was analysed extensively through physical, chemical, and rheological tests, and an ageing sensitivity index was employed to correlate their trends and provide a broader understanding of the ageing approaches. Further, the obtained results are compared with the field-aged RAPs collected from the same geographical region to assess the relevance of these ageing approaches under real field conditions.

2. Materials and Methods

2.1. Materials

In this study, a bitumen with a penetration grade of 50/70 was chosen considering the local availability and as the commonly used asphalt binder for the production of asphalt mixtures on the highways of the southernmost region of Italy. The loose asphalt mixtures were prepared using limestone aggregates according to the Italian technical specifications for the highways [27]. Furthermore, to have extensive knowledge regarding the correlation of the considered artificial ageing methods with field ageing mechanisms, reclaimed asphalt pavements (RAPs) from two different sources were included. RAP 1 was milled from the surface layer of a pavement with a known history of 12 years of service life and RAP 2 was from an unknown origin.

2.2. Accelerated Ageing Methods

The study consists of mainly two ageing approaches, one on the binder and another on the loose asphalt mixtures to simulate the long-term ageing mechanism of asphalt binder that occurs over the service life of a pavement. Figure 1 illustrates the comprehensive research plan adopted in this study. The extensive details of the ageing methods are described in the following sections.

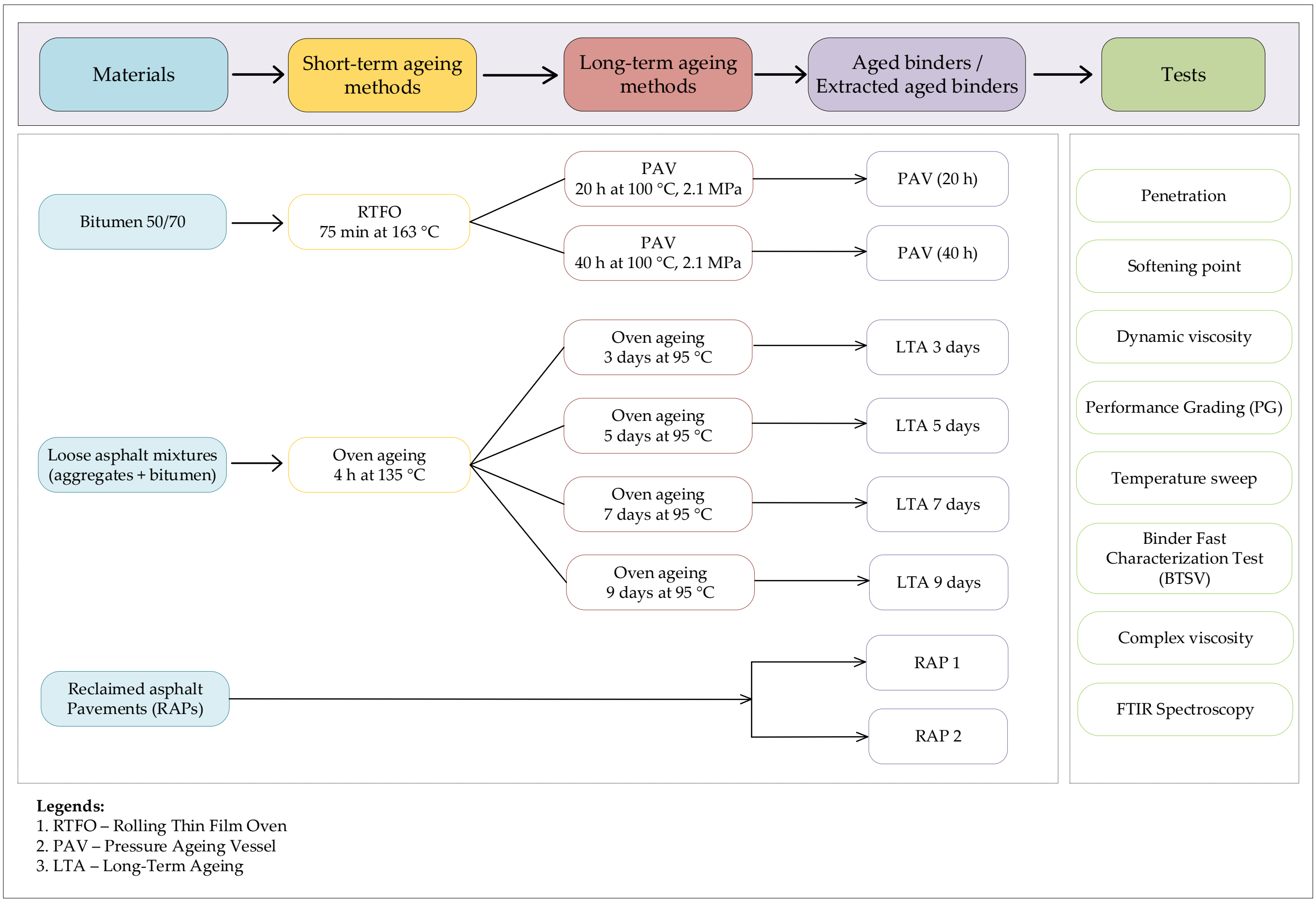

Figure 1. Research plan.

2.2.1. Ageing of Asphalt Binder

In this method, the asphalt binder was aged using the combination of a rolling thin film oven (RTFO) and a pressure ageing vessel (PAV) to simulate the long-term ageing mechanism. This approach was chosen since it is widely used and is the standard protocol for binder ageing. The RTFO is used to simulate the short-term ageing mechanism on asphalt binder in accordance with EN 12607-1:2014 [28]. For this purpose, 35 g of bitumen was placed in a glass bottle and kept in the RTFO setup for 75 min at a temperature of 163 °C with an airflow of 4 L/min. Subsequently, after the completion of short-term ageing using RTFO, the binder residual of 50 g was placed directly into each of the circular pans and was kept in the PAV for 20 h with an air pressure of 2.1 MPa according to EN 14769:2012 [29]. The ageing temperature of 100 °C was chosen based on the grade of the binder used in the study. Furthermore, to have extensive understanding of ageing behaviour with the increase in duration, the PAV-aged binder was repeatedly aged for an extra ageing duration of 20 h within the PAV under the same defined condition.

2.2.2. Ageing of Loose Asphalt Mixtures

In this approach, the loose asphalt mixtures were prepared according to an Italian technical specification [27] for a dense graded wearing course, and the mix design was optimised using the volumetric method. The grading distribution of the aggregates mix used to prepare the loose asphalt mixtures are summarised in Table 1. The binder content of 5% on the weight of the aggregates was determined as the optimum binder content to have an air void level of 4% on the compacted specimens at the design gyration level recommended in the technical specification. Based on this, all the loose asphalt mixtures were produced according to the defined mix design and were placed in a square tray

(460 mm × 460 mm × 50 mm) with a maintained height of 25 mm, which is shown in Figure 2. Subsequently, the loose asphalt mixtures were short-term-aged at 135 °C for 4 h, and afterwards, for the purpose of long-term ageing, the trays of asphalt mixtures were kept in a non-ventilated oven at 95 °C for various ageing durations of 3 days, 5 days, 7 days, and 9 days to investigate the ageing behaviour with respect to the increment in duration.

Table 1. Grading distribution of the loose asphalt mixture.

Sieve Size (mm)	Passing Percent (%)	
	Optimised Mix Gradation	Specification Limits
16	100	100–100
12.5	100	100–100
8	95.3	90–100
4	53.9	44–64
2	32.6	28–42
0.5	20.6	12–24
0.25	16.4	8–18
0.063	8.1	6–10

Figure 2. Loose asphalt mixture oven ageing.

2.3. Extraction and Recovery of Aged Binders

The extraction of binder from loose asphalt mixtures aged at discrete ageing durations and two RAP sources was carried out using standard binder extraction procedures prescribed in EN 12697-1:2020 [30]. The hot extractor method using a wire mesh was adopted and the solvent used for the extraction was tetrachloroethylene. A centrifuge was employed in order to remove the fine particles present in the extracted solution. Finally, the binder was recovered using a rotavapor according to EN 12697-3:2005 [31].

2.4. Testing Methods

2.4.1. Physical Properties

The physical properties of the aged binders were evaluated on the basis of traditional methods such as the penetration test, softening point test, and dynamic viscosity test. The penetration test was performed on unaged and aged binders at 25 °C to evaluate and compare the consistency of the binders. The temperature sensitivity of the asphalt binder aged using different ageing approaches was assessed via the softening point test. The dynamic viscosity test was conducted at various temperatures to investigate the flow and deformation behaviour of the aged binders.

2.4.2. Rheological Properties

The rheological characteristics of the binders aged using two ageing approaches and the field-aged binders were investigated using the Anton Paar MCR 102 dynamic shear rheometer (DSR). In this study, a temperature sweep test was performed within the temperature range of 25 to 90 °C to evaluate parameters such as the complex shear modulus (G*) and phase angle (δ). Furthermore, a comprehensive rheological analysis was carried out using test parameters including upper-performance grading, loss factor (tan δ), complex viscosity, and equi-shear modulus temperature and its corresponding phase angle.

2.4.3. Chemical Properties

The chemical characteristics of aged binders were examined using Fourier Transform Infrared (FTIR) spectroscopy. In this study, a Bruker Tensor 27 FTIR with an attenuated total reflectance (ATR) diamond crystal was employed to analyse the changes in the chemical functional group of the aged binders. This technique identifies chemical functionalities corresponding to distinct bonds or functional groups as a function of wavenumber by determining the infrared light absorbed by the bonds in molecules when its frequency matches the vibration frequency of the bonds [3]. The literatures showed that the spectral peaks corresponding to the wavelengths around 1030 cm^{-1} (sulfoxide group) and around 1700 cm^{-1} (carbonyl group) are significantly affected during the ageing process. The spectral peaks corresponding to the wavelengths around 1460 cm^{-1} and 1376 cm^{-1} are referred to as aliphatic groups, which are considered as the baseline for the analysis of a spectrum. The sulfoxide and carbonyl indices can be calculated using the formulae below [32].

$$\text{Sulfoxide Index(SI)} = \frac{\text{Area of the sulfoxide band centred at 1030 cm}^{-1}}{(\text{Area of the CH}_2 \text{ centred at 1460 cm}^{-1} + \text{Area of the CH}_3 \text{ centred at 1376 cm}^{-1})} \quad (1)$$

$$\text{Carbonyl Index(CI)} = \frac{\text{Area of the carbonyl band centred at 1700 cm}^{-1}}{(\text{Area of the CH}^2 \text{ centred at 1460 cm}^{-1} + \text{Area of the CH}^3 \text{ centred at 1376 cm}^{-1})} \quad (2)$$

3. Results and Discussion

3.1. Penetration Test

The consistency of the aged binders was assessed through a standard penetration test in accordance with EN 1426:2015 [33]. Figure 3 demonstrates the results of the penetration tests. Based on the results, all the aged binders, regardless of their ageing methods, exhibited a significant reduction in penetration value when compared to the virgin bit.50/70 and RTFO binder. The reduction in the penetration value clearly quantifies the effect of oxidative ageing on asphalt binder. The PAV (40 h) and LTA 9 days binders showed relatively lower and comparable penetration values despite the ageing techniques. This could provide a clear indication of the influence of ageing duration in terms of the consistency of the asphalt binder. Moreover, it was observed that the PAV (20 h) and LTA 5 days aged binders demonstrated a similar hardening effect. Comparing the penetration values of field-aged binders, RAP 1 correlated with the LTA 3 days, while RAP 2 showed results comparable to PAV (20 h), LTA 5 days, and LTA 7 days aged binders. Further, a previous study reported that two distinct RAP sources field-aged for 20 years exhibited the penetration values of 26.6 and 29.7 [34]. In comparison to these reported values, all the aged binders investigated in our study demonstrated lower penetration values during the analysis.

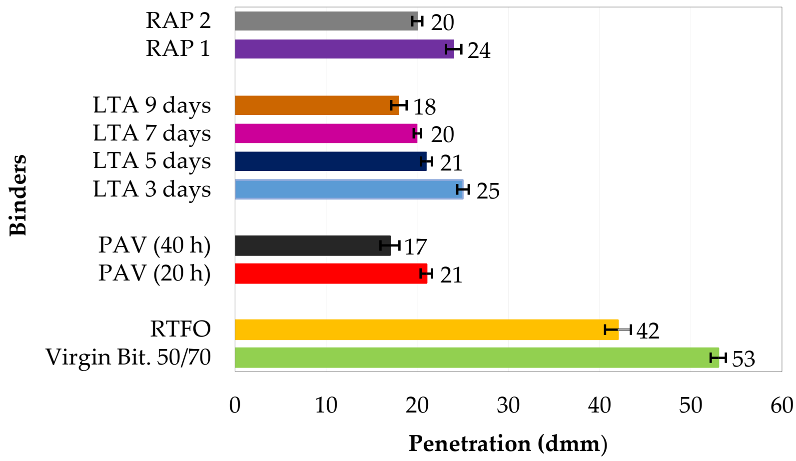

Figure 3. Penetration values.

3.2. Softening Point Test

In compliance with EN 1427:2015 [35], the thermal sensitivity of all the asphalt binders was determined through a softening point test using an automatic ring and ball tester. Figure 4 illustrates the softening point of all the investigated binders. The results clearly indicate that different ageing techniques influenced the softening point of the asphalt binder to different extents. Comparatively, the PAV (40 h) binder showed a higher softening point of 65.6 °C, which is 32.3% more than that of the unaged bitumen. It is worth mentioning that the PAV (20 h) and LTA 5 days binders exhibited similar softening point temperatures, which were nearly 22.6% higher than that of virgin bit.50/70. Nevertheless, the effect of the extensions in the ageing duration applied to the loose asphalt mixture could not exhibit comparable high temperature sensitivity with that of the PAV (40 h) aged binder. The softening point values of LTA 7 days and LTA 9 days showed a slight increase of 1.2% and 2.6% when compared to those of PAV (20 h) and LTA 5 days, respectively. The thermal conductivity of limestone aggregates could impact the degree of oxidation in the loose asphalt mixture approach, slowing down the rate of oxidation with subsequent increases in ageing durations. Considering the field-aged binders, the softening point of RAP 1 showed a similar value to LTA 7 days and had a relatable value to LTA 5 days and PAV (20 h). On the other hand, the results of RAP 2 demonstrated a comparable value to LTA 9 days. According to the literature [34], two RAP sources having an age of 20 years exhibited softening points of 61.4 and 61.7 °C, which are consistent with the values observed in this study for RAP 1.

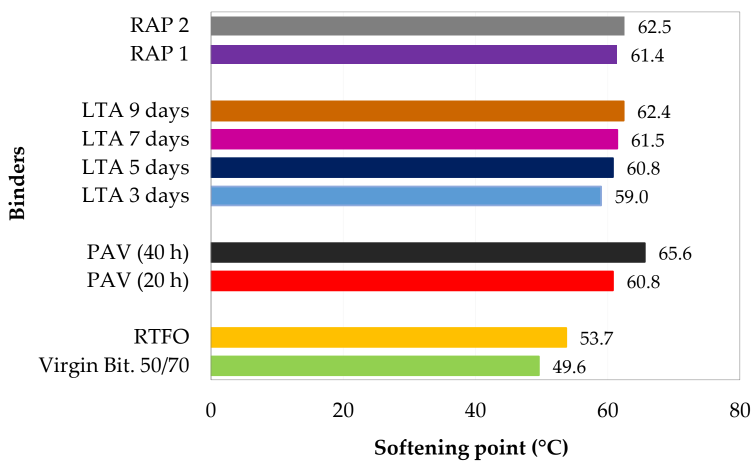

Figure 4. Softening point of the binders.

3.3. Dynamic Viscosity Test

The dynamic viscosity test was performed to measure the resistance to flow and deformation behaviour of the asphalt binders. For this purpose, a Brookfield rotational viscometer was used to conduct the dynamic viscosity test of the aged binders as per EN 13302:2018 [36]. In this research, a series of temperatures such as 135 °C, 150 °C, 160 °C, and 180 °C were considered to evaluate and compare the dynamic viscosity of the aged binders. The obtained results are presented in Figure 5. Based on the results, PAV (40 h) exhibited the highest viscosity across all the tested temperatures, followed by the LTA 9 days aged binder. All the extracted aged binders from the loose asphalt mixtures demonstrated a relatable viscosity property to PAV (20 h) at the tested temperatures and could not observed a similar trend of the ageing severity for PAV (40 h). However, it is worth mentioning that LTA 5 days and PAV (20 h) showed a comparable dynamic viscosity at all the performed temperatures. In view of the field-aged binders, both RAP 1 and RAP 2 demonstrated lower dynamic viscosity at all the tested temperatures compared to PAV (40 h).

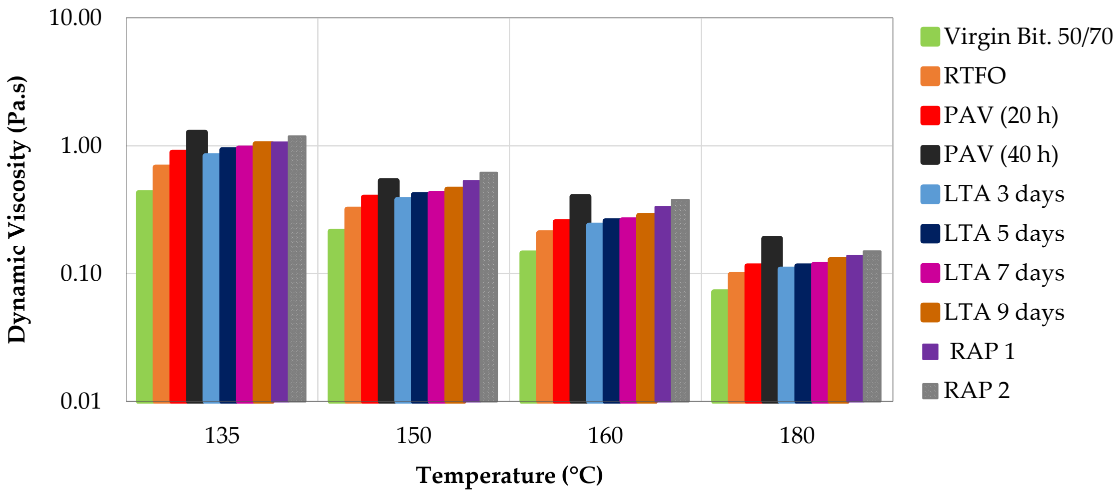

Figure 5. Dynamic viscosity of the aged binders over various temperatures.

3.4. Performance Grading (PG)

The upper-performance grading of all the aged binders was determined using a DSR. In this study, the test was performed at a frequency of 1.59 Hz using a 25 mm plate geometry and a 1 mm gap in accordance with EN 14770:2012 [37]. The test started at 46 °C, and the temperature gradually increased at a rate of 6 °C until the test met the criterion of $G^*/\text{Sin}\ \delta$, which equals 1 kPa. The same test conditions were employed for all the aged binders and the virgin bitumen, considering the main objective of the study to assess the ageing correlations. It is expected that during the ageing of binders, their complex shear modulus (G^*) is supposed to increase with the decrease in the phase angle (δ) and thus the stiffness of the binder increases. Consequently, the asphalt binder aged under various conditions and ageing methods depicts variation in the PG values at high temperatures. The higher degree of the ageing level can be related to a higher value in the upper PG. According to the results, summarised in Table 2, the effect of ageing can be clearly observed by the increase in the upper PG values in comparison with virgin bitumen. For the purpose of comparison, the upper critical temperature was mainly considered in the study whereas the standard upper PG of the aged binders was found to be similar. The upper critical temperature refers to the failure temperature at which the test complies and $G^*/\text{Sin}\ \delta$ equals to 1 kPa. It is noteworthy that among all the studied ageing methods, the PG and the upper critical temperature of the PAV (40 h) showed the highest value. The LTA 9 days showed the highest upper critical temperature among the loose asphalt mixture ageing methods. However, the ageing mechanism of loose asphalt mixture showed a deceleration in the ageing level with an increase in the duration after reaching a particular ageing state compared to the binder ageing technique. The PAV (40 h) depicted an increase of 6.5% and 26.2% of upper critical temperature compared to the PAV (20 h) and the virgin bit. 50/70, respectively. On the other hand, considering the loose asphalt mixture ageing technique, the LTA 5 days demonstrated a similar PG and upper critical temperature compared to the PAV (20 h). The binder LTA 9 days showed 2.3% and 21.2% increase in upper critical temperature compared to LTA 5 days and virgin bit. 50/70, respectively. Accordingly, it is worth mentioning that the aged binders, PAV (20 h) and the LTA 5 days demonstrated similar PG and upper critical temperature during the analysis. Meanwhile, the field-aged binder RAP 1 showed a comparable result with LTA 5 days, LTA 7 days, and PAV (20 h). The results of RAP 2 were relatable to LTA 9 days, and both the field-aged binders exhibited a relatively lower critical temperature than PAV (40 h).

Table 2. Results of the upper performance grading.

Binder	Original Upper PG (°C)	Upper Critical Temperature (°C)
Virgin Bit. 50/70	64	67.5
RTFO	70	72.0
PAV (20 h)	76	80.1
PAV (40 h)	82	85.2
LTA 3 days	76	78.4
LTA 5 days	76	80.0
LTA 7 days	76	80.6
LTA 9 days	76	81.8
RAP 1	76	80.9
RAP 2	82	82.4

3.5. Temperature Sweep Test

The temperature sweep test was performed on a dynamic shear rheometer in order to investigate the behaviour of all the aged binders and virgin bitumen within the temperature range of 25 °C to 90 °C. The test was carried out on a plate geometry of 25 mm with a gap of 1 mm between the plates and a constant shear stress of 500 Pa at a frequency of 10 rad/s with an increase in temperature of 1 °C/min. The results of the test are depicted in Figures 6 and 7, which are in terms of the loss factor and the isochronal plot of the complex shear modulus (G*), respectively. The loss factor of the asphalt binder is determined by the loss modulus (G'') and the storage modulus (G'). The G'' indicates the lost deformation energy of the material and serves as the viscous component of the material, whereas the G' represents the energy stored in the system and reflects the elastic part of the material. The parameter G'' is directly proportional to the loss factor that often depicts material losses, and which indicates the softness or hardness of the asphalt binder [3]. Since G'–G'' equilibrium indicates the structure of a material, a rheological investigation employing this approach is regarded as valid for evaluating the behaviour of the ageing of asphalt binders. The loss factor can be calculated as the ratio of G'' to G'.

In accordance with the obtained results, it is obvious that all the ageing methods increased the stiffness of the binder collated to the virgin bit. 50/70. The lower value of the loss factor and higher value of the complex shear modulus of the asphalt binder represent its higher stiffness property. Therefore, based on different ageing approaches, the PAV (40 h) aged binder showed the highest ageing level, which can be observed in the loss factor and isochronal plots of G*. It is noteworthy that the LTA 5 days binder and the PAV (20 h) aged binders depicted similar curves in both plots, which indicates that a similar stiffness effect occurred during both ageing approaches. Among the other loose asphalt mixture aged binders, LTA 7 days aged binder demonstrated a curve very close to that of PAV (20 h) and LTA 5 days. It is worth mentioning that the stiffness of LTA 9 days aged binder increased compared with other loose mixture aged binders; however, the intensity of the ageing effect in terms of stiffness remains less compared to PAV (40 h). The loss factor and isochronal curves of the field-aged binder RAP 1 were comparable with LTA 5 days, PAV

(20 h), and LTA 7 days. While the isochronal curve of RAP 2 exhibited a similar trend with LTA 9 days and during the loss factor analysis, it was observed that the loss factor of RAP 2 comparatively decreased after 60 °C with that of LTA 9 days.

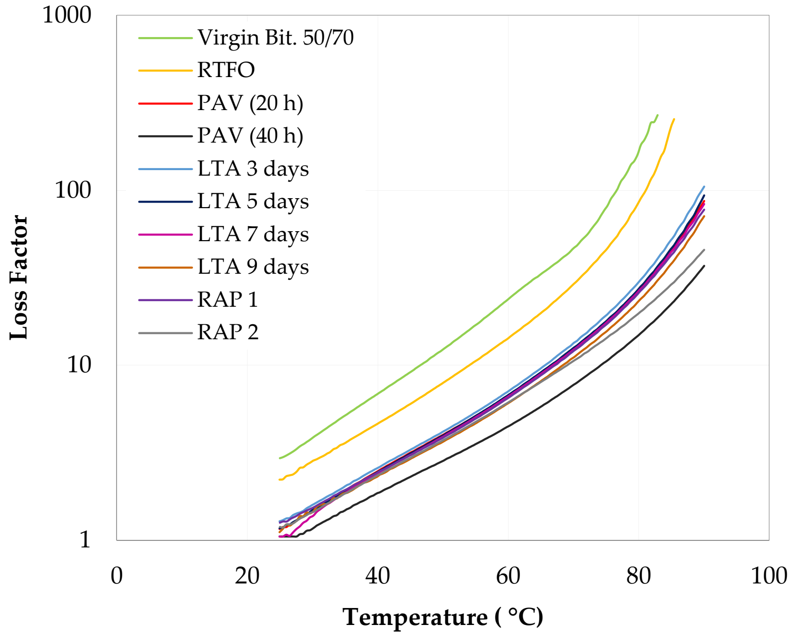

Figure 6. Loss factor versus temperature curve.

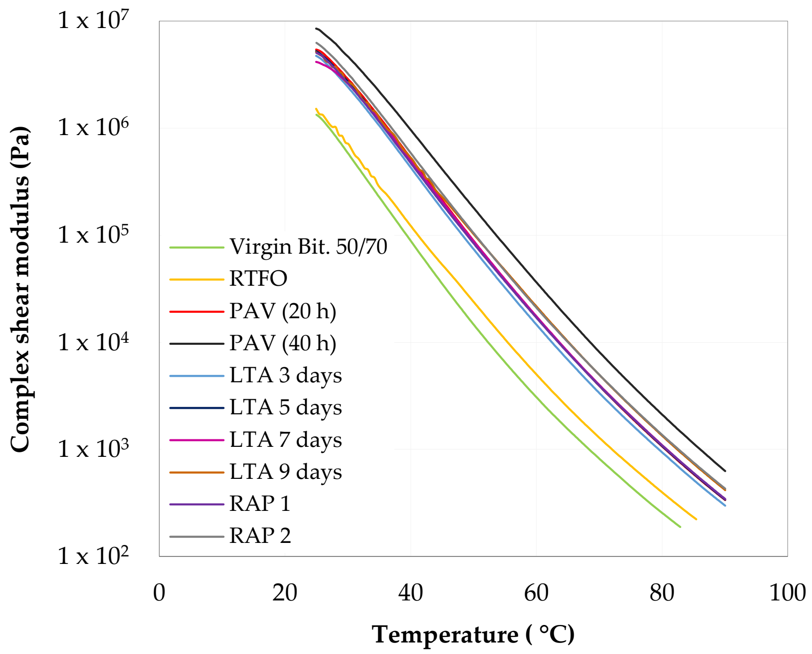

Figure 7. Isochronal plots of the complex shear modulus (G*).

3.6. Binder Fast Characterisation Test (BTSV)

The BTSV test was used to evaluate the deformation behaviour of the asphalt binder at high temperatures using a DSR. The test determined the equi-shear modulus temperature (T_{BTSV}) and the corresponding phase angle (δ_{BTSV}) at which the asphalt binder exhibited a complex shear modulus (G*) of 15 kPa under a stress-controlled oscillation mode at a gradually increasing temperature as per EN 17643:2022 [38]. The viscoelastic properties of asphalt binders can be investigated depending on the equi-shear modulus temperature, which correlates with the hardness of the binder and their corresponding phase angle to assess the degree of alteration. In this study, the following test settings were adopted:

- Plate geometry: 25 mm;
- Gap: 1 mm;
- Constant shear stress: 500 ± 5 Pa;
- Test frequency: 1.59 Hz;
- Temperature range: 25 °C to 90 °C;
- Temperature increase rate: 1.2 °C/min.

The results of the BTSV test are summarised in Table 3. According to the obtained results, the equi-shear modulus temperature of all the aged binders increased and their corresponding phase angle decreased compared to that of virgin bit.50/70 and RTFO aged binder. Significantly, PAV (40 h) binder showed the highest equi-shear modulus temperature (T_{BTSV}) and lowest phase angle (δ_{BTSV}) among other aged binders. However, different ageing approaches steered to different ageing levels of the binders. It is noteworthy that the results of the BTSV test also affirm a similar trend between PAV (20 h) and LTA 5 days aged binders, which demonstrated a comparable T_{BTSV} and δ_{BTSV}. The LTA 7 days and LTA 9 days aged binders showed a slight increase in the test parameters compared to LTA 5 days and PAV binders. Nevertheless, all the binders demonstrated comparable T_{BTSV} values in contrast to their respective softening point temperature.

Table 3. Results of the binder fast characterisation test.

Binder	T_{BTSV} (°C)	δ_{BTSV} (°)
Virgin Bit. 50/70	49.9	85.3
RTFO	52.9	83.9
PAV (20 h)	61.0	81.9
PAV (40 h)	65.8	80.6
LTA 3 days	59.8	82.3
LTA 5 days	60.9	81.9
LTA 7 days	61.4	81.8
LTA 9 days	62.4	81.6
RAP 1	61.5	81.7
RAP 2	62.3	81.5

3.7. Complex Viscosity

The complex viscosity is a parameter, which can be calculated as the ratio of the complex shear modulus to their corresponding angular frequency. Figure 8 illustrates the complex viscosity curve of the aged binders within the temperature range of 25 to 90 °C. It is evident that all the long-term aged binders, despite their ageing methods, exhibited high complex viscosity compared to virgin bit. 50/70 and RTFO. In order to compare the effect of ageing, the complex viscosity at 60 °C was considered on the basis of its significance in the performance evaluation. It is worth mentioning that the PAV (20 h), LTA 5 days, and LTA 7 days exhibited comparatively similar and nearly 5.6 times higher viscosity than that of the virgin bit. 50/70. It was observed that PAV (40 h) showed the highest complex viscosity at 60 °C, which is almost 12 times higher than that of the virgin bitumen. The extended duration of 20 h with the combination of induced pressure of 2.1 MPa and temperature of 100 °C after the standard PAV could significantly escalate the degree of ageing, which results in a high stiffness of the binder. The complex viscosity of LTA 3 days and LTA 9 days demonstrated 4.8 times and 7 times higher than that of the virgin bit. 50/70, respectively. These results demonstrate that long-term ageing methods on loose asphalt mixtures could not drastically increase the degree of oxidative ageing similar to that of PAV (40 h) by extending the ageing duration. It was observed that the complex viscosity curve of RAP 1 was comparable with LTA 5 days, LTA 7 days and PAV (20 h). On the other hand, RAP 2 exhibited a relatable curve with the LTA 9 days aged binder.

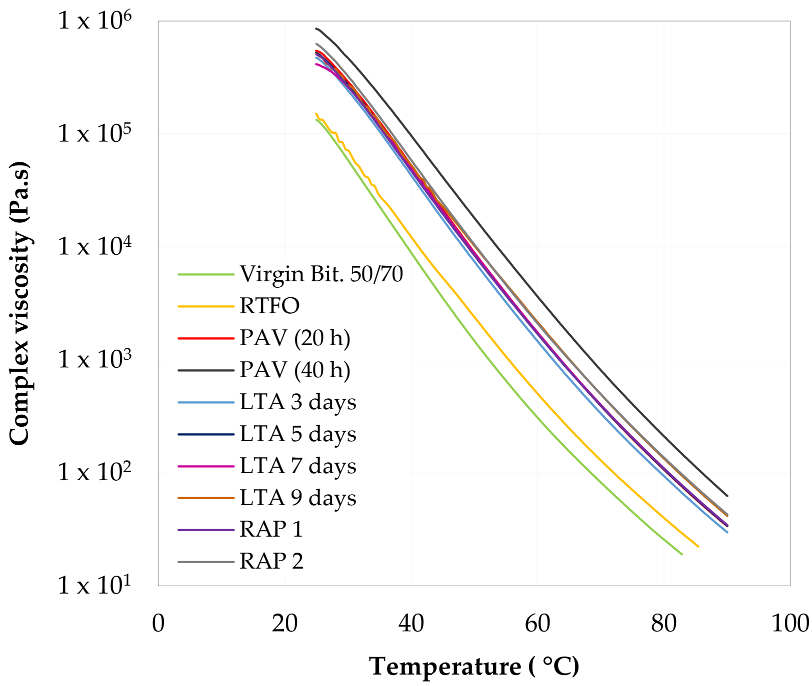

Figure 8. Complex viscosity curve.

3.8. Fourier Transform Infrared (FTIR) Spectroscopy

In this study, an ATR FTIR spectroscopy was used to evaluate the chemical changes in the functional groups of the asphalt binder that occur during different ageing methods. For this purpose, the wavelengths of the spectral peaks around 1030 cm^{-1} (sulfoxide group) and around 1700 cm^{-1} (carbonyl group) were analysed as they are the spectral peaks of the functional groups significantly affected during the ageing of asphalt binder which were reported in many studies. Figure 9 shows the average obtained spectra of the aged binders and the virgin bit. 50/70. The spectral peaks corresponding to the sulfoxide band and carbonyl bands of all the aged binders increased during the oxidation phase compared to the virgin bit. 50/70. It is worth mentioning that the spectral peaks of PAV (40 h) were the highest among the aged binders followed by PAV (20 h). A similar intensified trend was not observed among other long-term aged binders. However, the oxidative ageing could be identified by the increase in the peaks of sulfoxide and carbonyl bands. This was also observed in the case of field-aged binders. The calculated sulfoxide and carbonyl indices are summarised in Table 4. According to the calculated indices, the binders aged using PAV exhibited higher indices than the loose mixture ageing method carried out in the oven. Overall, it can be deduced that the application of air pressure during the PAV ageing method accelerated the chemical changes in the significant functional groups in the bitumen. In contrast, due to the absence of air pressure and the influence of the mineral aggregate composition in the loose asphalt mixture ageing method, the intensity of the chemical changes was comparatively slower than that of the binder ageing method. The spectrum analysis provided clear knowledge on the impact of ageing in the chemical functional group; however, a comparable trend could not be observed with this method. The calculated indices of RAP 1 and RAP 2 showed less ageing severity compared to PAV (20 h) and PAV (40 h). For instance, the severity of PAV (20 h) and PAV (40 h) was 1.2 and 1.6 times greater, respectively, than that of the investigated field-aged RAPs. The sulfoxide indices of both RAP binders exhibited comparable values to those of aged binders from loose asphalt mixtures, regardless of the ageing durations.

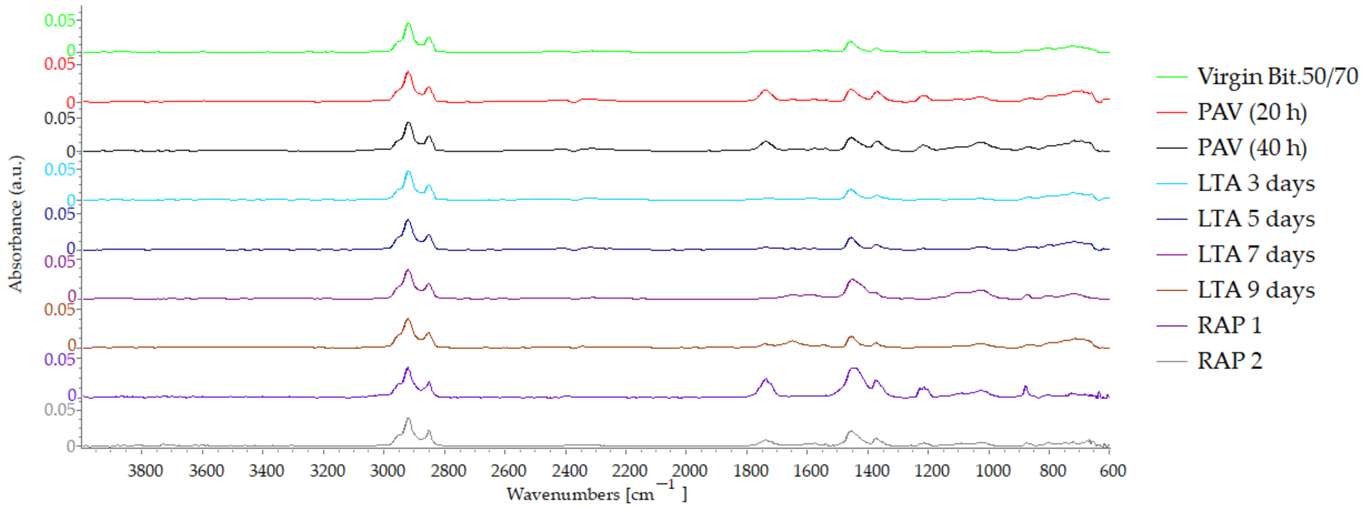

Figure 9. Average obtained spectra of the investigated binders.

Table 4. Calculated average values of sulfoxide and carbonyl indices.

Binder	Sulfoxide Index (SI)	Carbonyl Index (CI)
Virgin Bit. 50/70	0.12	0.08
PAV (20 h)	0.31	0.48
PAV (40 h)	0.58	0.54
LTA 3 days	0.20	0.17
LTA 5 days	0.24	0.23
LTA 7 days	0.25	0.26
LTA 9 days	0.27	0.31
RAP 1	0.24	0.45
RAP 2	0.27	0.40

3.9. Comparative Evaluation Based on the Ageing Sensitivity Index

The aim of this study was to investigate and compare the effect of long-term ageing sensitivity of asphalt binder using two thermo-oxidative ageing approaches. More specifically, based on the results of the performed tests, it was shown that both ageing approaches were able to accelerate the ageing to a certain extent. In order to meticulosusly evaluate the intensity of the investigated ageing methods, an ageing sensitivity index was deployed. This can be used to assess the severity of ageing with respect to its original unaged properties of the asphalt binder. This index tool can be utilised to identify the ageing sensitivity of each ageing approach with the increase in ageing duration. The ageing sensitivity index can be calculated according to the following equation:

$$\text{Ageing Sensitivity Index} = \frac{|(\text{Evaluated parameter after ageing} - \text{Evaluated parameter before ageing})|}{\text{Evaluated parameter before ageing}} \tag{3}$$

Figures 10 and 11 depict the calculated ageing sensitivity index for all the assessed parameters. The increase in the ageing sensitivity index signifies the severity of ageing with respect to the unaged condition and its resultant impact on each test parameter. Based on the observed results, among the various ageing methods examined, PAV (40 h) demonstrated the highest ageing sensitivity index. It is worth noting that the standard binder ageing protocol, PAV (20 h), and the loose asphalt mixture aged for 5 days exhibited a very similar ageing index during the rheological evaluation. Significantly, it is noted that the degree of ageing of the loose asphalt mixture ageing approach was slower as the ageing duration increased. According to the literature, the surface of the aggregates can adsorb the most polar compounds of bitumen in the fractions of asphaltenes and resins [39,40]. The adsorption, along with the thermal conductivity of limestone aggregates, could decelerate the oxygen diffusion in the studied loose asphalt mixtures. On the other hand, extending the PAV duration to 40 h considerably accelerated the ageing level due to the repeated application of 2.1 MPa pressure and a temperature of 100 °C. It is essential to highlight that such an extension in ageing duration did not align with the ageing of the studied field-aged RAP binders in both rheological and chemical analysis. Consequently, the extension of PAV duration demonstrated a more pronounced ageing effect compared to all the investigated binders. Considering the sum of all the calculated ageing sensitivity indices for the physical and rheological parameters, PAV (40 h) showed 2.1 times the severity of PAV (20 h) and 1.7 times with LTA 9 days. Since data on the unaged properties of field-aged binders were unavailable, RAP binders were excluded from this specific analysis. In the chemical analysis, the carbonyl and sulfoxide indices of PAV (20 h) and PAV (40 h) were relatively higher than those of the aged loose asphalt mixtures, indicating a high sensitivity to ageing among the investigated aged binders. In particular, LTA 5 days and PAV (20 h) showed mostly identical results of the physical and rheological analyses. The FTIR spectroscopy analysis, on the other hand, demonstrated that the ageing sensitivity index for the chemical parameters of PAV (20 h) was 2.3 times greater than that of LTA 5 days, indicating a varying trend among the rheological and chemical behaviours of the asphalt binder.

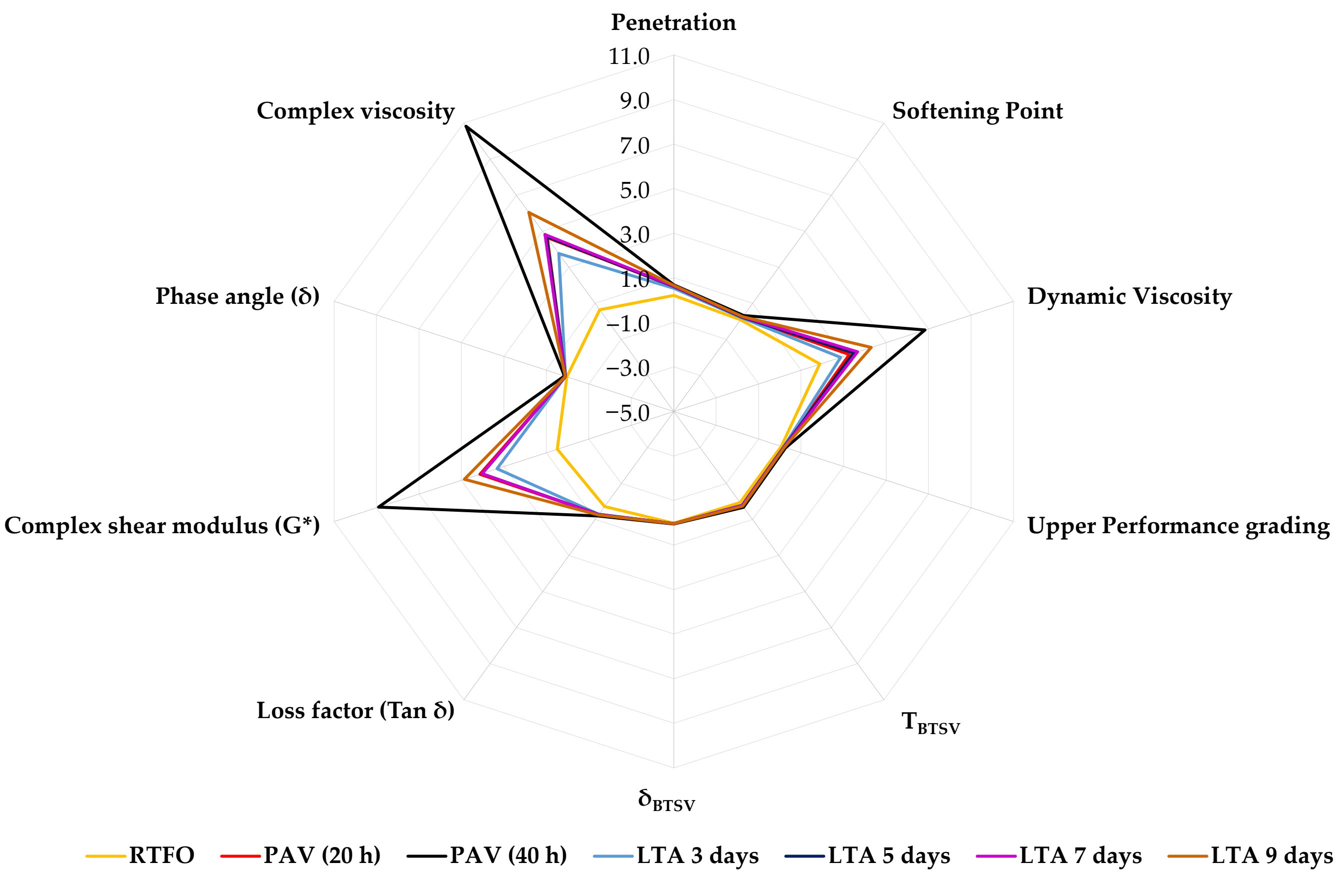

Figure 10. Ageing sensitivity index of the investigated physical and rheological parameters.

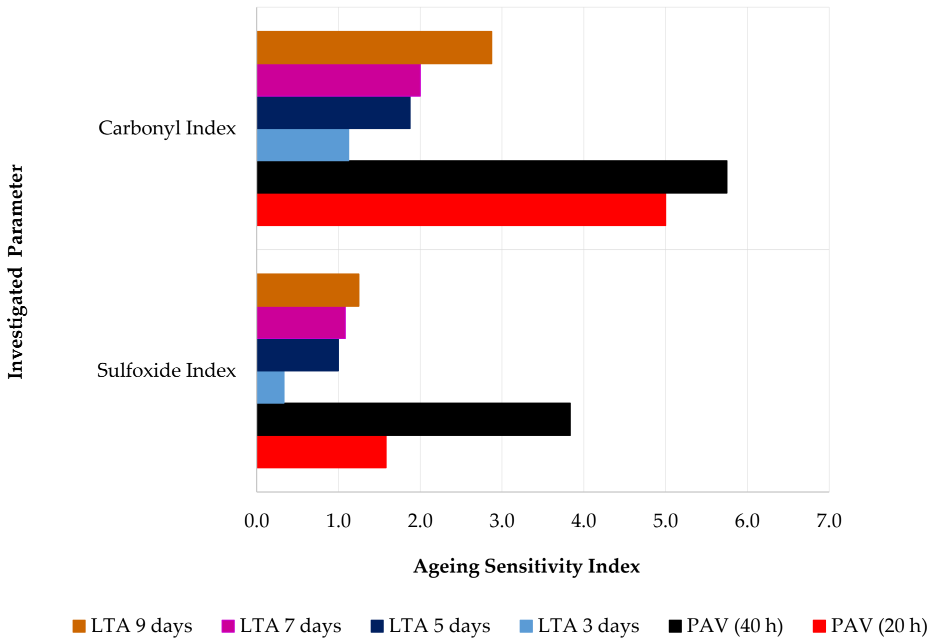

Figure 11. Ageing sensitivity index of the investigated chemical parameters.

4. Summary and Conclusions

This study addressed a comprehensive evaluation of the effect of thermo-oxidative ageing methods on asphalt binder, their correlation, and their relevance to field-aged binders. For this purpose, two artificial ageing approaches were employed in the study, one on the binder and another on the loose asphalt mixtures with distinct ageing durations. The impact of these ageing methods on the behaviour of asphalt binder was investigated using physical, rheological, and chemical characterisation. The ageing degree caused by these ageing methods was meticulously evaluated using an ageing sensitivity index.

Based on the results, it was observed that both ageing approaches for the asphalt binder could significantly accelerate the ageing degree to a certain level with an increase in the duration. The ageing sensitivity analysis signified that PAV (20 h), which is the standard binder ageing protocol, and the loose asphalt mixture aged for 5 days demonstrated a very similar ageing trend among the investigated rheological parameters. Alongside that, the intensity of oxidative ageing in the loose asphalt mixture decreased after a certain level of ageing, and further increments in the ageing duration did not escalate ageing severity compared to that of PAV ageing. The adsorption of the polar compounds of bitumen on the aggregate surface, along with the thermal conductivity of limestone aggregates, could account for decelerating the oxygen diffusion in the loose asphalt mixtures. In this regard, a decline in the oxidation rate was noticed in the cases of the LTA 7 days and LTA 9 days aged binders. The highest ageing degree was observed in the PAV (40 h) aged binder, which was higher than that of all the studied approaches and field-aged binders, showing the severity of such an ageing procedure. On the other hand, the rheological and chemical properties of the loose asphalt mixture ageing method provided an analogous relationship with the studied field-aged RAPs. However, these observed trends might not be evident enough to predict the ageing degree of certain years of field ageing phenomena and require further investigation. The FTIR spectroscopy analysis revealed that the ageing methods influenced the functional groups associated with ageing in distinct ways, even though they exhibited similar rheological behaviour. The studied chemical indices of binder aged in the PAV for 20 and 40 h exhibited significantly high values, which were not consistent with the field-aged binders.

Overall, it can be inferred that this study could yield significant insights into various accelerated ageing methods for asphalt binders and their correlation with the standard ageing protocol. Thermo-oxidative ageing methods are consistently employed to assess the long-term performance and ageing characteristics of asphalt binders, whether they include modifiers or not. Contrastingly, several factors influence the ageing behaviour of asphalt pavement, oxidative ageing remains a crucial aspect. Based on the results, it becomes evident that in order to evaluate the ageing behaviour or long-term performance of any asphalt binder, it is essential to consider the thermal properties of the mineral aggregates, as well as their composition within the mixture, to gain a comprehensive understanding. Additionally, due to the absence of a standardised ageing protocol for loose asphalt mixtures, this study opens significant possibilities for comprehending the ageing mechanisms within these asphalt mixtures. This comparative assessment of the loose asphalt mixture ageing protocol could lead to a way to implement this approach to evaluate the ageing mechanism and long-term performance of dry method modification in asphalt mixtures, which was considerably challenging to evaluate. It provides a cohesive evaluation of the expected life cycle of a binder and thus allows us to understand the service life and the time window of the life cycle of a binder due to its ageing. However, the presented results are only valid for the specified type of bitumen, and further investigation is recommended with modified binders and other mineral aggregates in loose mixture ageing to gain a prolonged knowledge of the ageing mechanism and its associated mixture-level performance. Further study on the influence of UV rays, along with thermo-oxidative ageing, of loose asphalt mixtures is also proposed and underway.

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Conflicts of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as potential conflict of interest.

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