

Review

Aging evolution and sustainability implications of crumb rubberized asphalt binder: A state-of-the-art

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ABSTRACT

Crumb rubberized asphalt binder achieves value-added recycling of waste scrap tires, as well as enhancement of mechanical properties and aging resistance of asphalt pavements. This paper provides a systematic literature review of the aging evolution and sustainability implications of crumb rubberized asphalt binder based on peer reviewed publications during 1993–2022. The analysis of the literature shows that aging methods, chemical-microstructural characterization, rheological-mechanical properties, and environmental benefits are the main research directions in studying aging of crumb rubberized asphalt binder. Furthermore, recent investigations mainly concentrate on the thermal aging behavior of crumb rubberized asphalt binder, while ultraviolet-induced aging and weathering aging lack attention. Rubber degradation is a characteristic manifestation of crumb rubberized asphalt binder during aging that can be characterized by chemical investigations. Aging-induced cracks are mitigated by rubber particles shown by microstructural observations. Rheological-mechanical investigations show different evolutions in high-temperature properties of crumb rubberized asphalt binder after aging, but a superior low-temperature cracking resistance as compared to base asphalt as well as styrene-butadiene-styrene modified asphalt has been observed. The inconsistent findings in the fatigue properties of aged crumb rubberized asphalt binder are compared and attributed to the selection of evaluation indicators. Life cycle assessment shows that depending on the local waste management policy, significant environmental benefits of crumb rubberized asphalt binder in terms of reduced energy consumption and global warming potential are possible.

1. Introduction

The increasing number of vehicles brings a heavy environmental burden, of which the disposal of end-of-life- tires has become a critical issue in recent decades. Almost 1 billion end-of-life-of tires are produced worldwide every year, accounting for nearly 2% of the total solid waste (Karaağaç et al., 2017). This abundant solid waste flow is also considered as valuable and attractive urban mining, which is already used as fuel or recycled in civil engineering materials (Dabic-Miletic et al., 2021). In the field of pavement infrastructure, end-of-life-of tires are usually mechanically ground into crumb rubber and mixed with asphalt materials as a performance-enhancing additive. Herein, “wet process” and “dry process” are the two major applications of crumb rubber as an

asphalt additive: “wet process” refers to adding the crumb rubber to the asphalt binder to produce the so-called crumb rubberized asphalt (CRM); “dry process” is using crumb rubber directly mixed with the asphalt mixtures (Picado-Santos et al., 2020). Since the invention by Charles McDonald in the middle of the 1960s, CRM produced by the wet process has shown its significant benefits in the mechanical properties and environment, continuously attracting the interests of researchers and engineers (Way, 2012). Nevertheless, as the service life of asphalt pavements increases, CRM demonstrates the concerning field performance when subjected to the practice environment (Osmari et al., 2019). The dry process is gaining more attention, as from a practical point of view it is easier to implement (Bueno et al., 2021).

Asphalt binder is exposed to multi environmental factors during its

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service life, including oxygen, heat, ultraviolet (UV) light, and humidity (Petersen, 2009; Poulidakos et al., 2014; Yuan et al., 2023). These complex environmental factors and repeated vehicle loading cause the aging of asphalt binder, thereby contributing to the deterioration of asphalt pavement (Jiang et al., 2022; Tauste et al., 2018). The aging behavior of base asphalt binder has been deeply investigated, showing the agglomeration of asphaltene molecules and the depletion of light fractions in asphalt binder as the dominant changes during aging (Petersen and Harnsberger, 1998). Agglomeration refers to the formation of larger asphaltene agglomerates with strong intermolecular interactions, due to the increasing concentration of polar functional groups during aging (Pahlavan et al., 2019). Depletion of light fractions refers to the evaporation of volatile components (saturates and aromatics) during aging, which decreases the flexibility of the binder (Apeagyei, 2011). Meanwhile, the decreased saturates and aromatics contents also results in the destabilization of the asphaltenes, thereby intensifying their agglomeration. These aging-related physicochemical changes increase the viscosity and brittleness of asphalt binder, subsequently inducing low-temperature and fatigue cracking (Cavalli et al., 2018; Petersen and Glaser, 2011). The aging mechanism of asphalt binder indeed lays a fundamental framework to understand the aging process of CRM but cannot fully unravel the physicochemical changes and performance evolution.

Due to the significant effect of aging on the properties of asphalt pavement, many studies have focused on the aging of CRM from different perspectives, including chemical changes, molecular size distribution, and rheological properties. Therefore, the current paper provides a technical overview of the aging mechanism and performance evolution of CRM. This study aims to discuss different aging factors and evaluation indicators on the aging properties of CRM, providing comprehensive insights from the existing studies.

2. Methodology

A thorough and systematic review of the literature is an essential feature of any academic project, creating a firm foundation for advancing knowledge, facilitating theory development where a plethora of research exists, and uncovering areas where research is needed (Webster and Watson, 2002). A Systematic Literature Review (SLR) is a reliable approach to mapping and assessing the relevant intellectual territory in order to answer related questions to the topic of interest and provide opportunities for future research (Goodhue and Thompson, 1995). SLR was conducted to evaluate the body of literature on the characterization methods of the aging and rejuvenation of asphalt binders, employing the five-step structure that included: (1) problem and topic definition; (2) selection of databases; (3) literature selection;

(4) critical appraisal and evaluation of selected studies; (5) synthesis and review writing. The steps of the systematic review are detailed in supplementary material.

3. Evaluation of publication focus

This section summarized the research tendency and interests in the aging of crumb rubberized asphalt based on the collected documents. The publication amounts and sources are shown in Fig. 1, indicating that the earliest publication in this filed is from 1993. Even though crumb rubber was used as an asphalt modifier from the early 1960s (Scofield, 1989), the aging of crumb rubberized asphalt started attracting research interest 30 years later and the increasing trend appeared since the year of 2016. This indicates more research interests would be drawn in the future due to the more deployment of crumb rubberized asphalt and their increasing service years.

To distinguish the research dimensions, the abstracts of collected 560 papers are analyzed using “Bibliometrix” software for calculating the occurrences of each word in abstracts per year (Aria and Cuccurullo, 2017). The top 100 high-frequency words were subsequently reviewed manually to remove the irrelevant words (materials name, searching keywords, and preparation methods), after which similar words were merged. The occurrence numbers of high-frequency words in abstracts per year were drawn in Fig. 2, showing the main research focus in this field. All high-frequency words can be divided into three research dimensions: aging methods (thermal aging and UV aging), chemical-microstructural characterization (Fourier-transform infrared spectroscopy, gel permeation chromatography, scanning electron microscope, and atomic force microscopy), and rheological-mechanical properties (rheological properties, low temperature properties, aging resistance, fatigue properties, high temperature properties, bending beam rheometer, and moisture damage). Furthermore, increasing publications mentioned the topics of Environmental Product Declaration (EPD) in the abstracts or keywords over recent 5 years as shown in Fig. 2, showing a novel research field.

Aging methods (thermal aging and UV aging) attracts the highest research interest in the selected publications, discussing the laboratory aging methods, sample preparation, and the effects of different aging factors. The chemical analysis (Fourier-transform infrared spectroscopy and gel permeation chromatography) is adopted to investigate the composition changes of crumb rubberized asphalt binder during aging. Meanwhile, advanced microscopic characterizations (scanning electron microscope and atomic force microscopy) attract rapidly increasing interest to observe the microstructures of crumb rubberized asphalt during aging. The rheological property of asphalt binder is a perpetual topic, which is also widely investigated for the aged crumb rubberized asphalt.

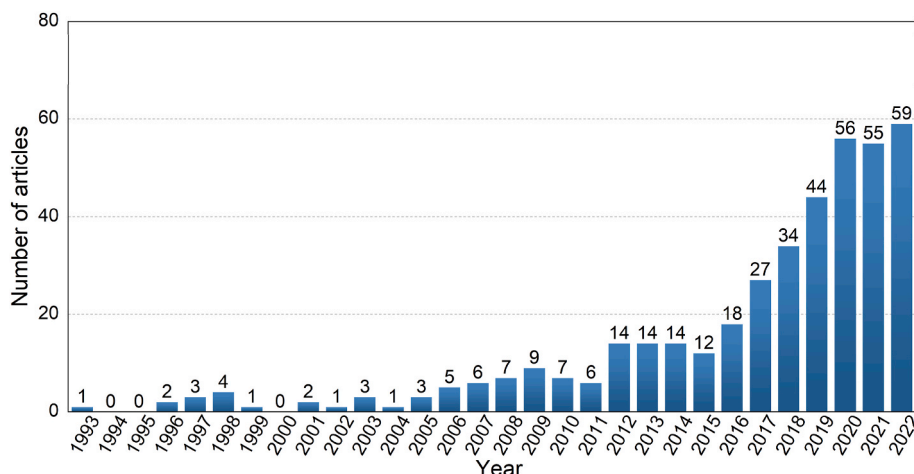


Fig. 1. Publication numbers per year on the topic of “asphalt (bitumen), rubber, and aging”.

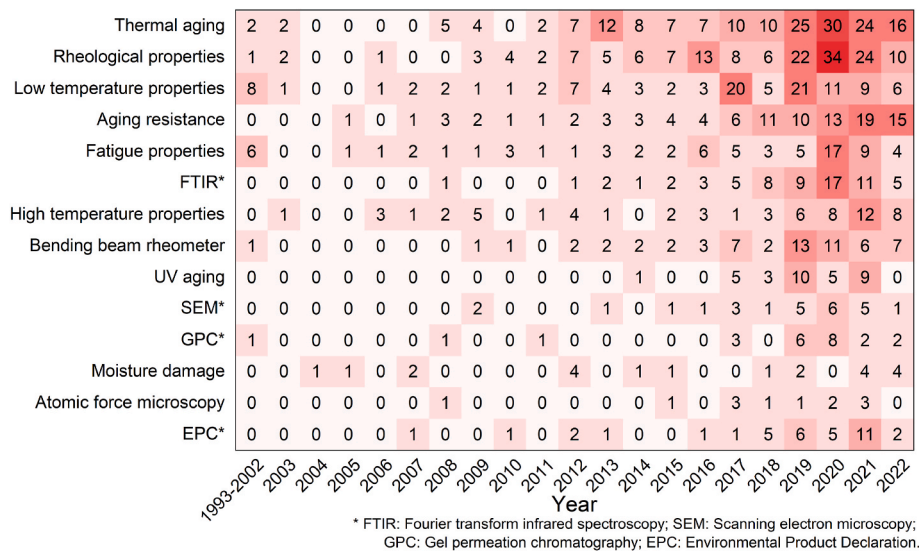


Fig. 2. Occurrences of high-frequency key words per year in abstract and keywords.

Specifically, the aging resistance and high temperature properties are mainly evaluated based on rheological experiments. Therefore, the publications focusing on rheological properties are discussed along with aging resistance, and high temperature properties. The low temperature properties (including “bending beam rheometer”), fatigue properties, and moisture damage are also widely investigated for the aged crumb rubberized asphalt, which is discussed in Section 6.2, 6.3, and 6.4, respectively. The publications on EPD are discussed in Section 7.

4. Aging methods

Asphalt aging is a complex and unavoidable physicochemical process during the use phase, which is mainly caused by the oxygen, heat, and UV light (Airey, 2003; He et al., 2023). To simulate the aging effects of various environmental factors, several laboratory aging methods are adopted to obtain the aged crumb rubberized asphalt binder, showing their different impacts on the rheological properties and chemical compositions (Poulikakos et al., 2019). This section presents the various aging methods used for the crumb rubberized asphalt binder. Considering the existing widely used standard thermal aging procedures, the non-standard aging methods are discussed in detail to provide insight into the laboratory aging design for crumb rubberized asphalt binder.

4.1. Thermal aging

Laboratory thermal aging is a common method to obtain the aged crumb rubberized asphalt binder, which includes the short-term aging and long-term aging. For short-term aging, it simulates the aging behavior of the asphalt binder during production, mixing, and paving by the thin-film oven test (TFOT, ASTM D1754) (ASTM D1754. *Standard Test Method for Effect of Heat and Air on Asphaltic Materials (Thin-Film Oven Test)*, 2010) and the rolling thin film oven test (RTFOT, ASTM D2872) (ASTM D2872. *Standard Test Method for Effect of Heat and Air on a Moving Film of Asphalt Binder (Rolling Thin-Film Oven Test)*, 2022). The pressurized aging vessel (PAV, ASTM D6521) (ASTM D6521. *Standard Practice for Accelerated Aging of Asphalt Binder Using a Pressurized Aging Vessel (PAV)*, 2022) is used to simulate the long-term aging of asphalt binder during the use phase of pavement. Meanwhile, several revised laboratory thermal aging methods are also conducted on the crumb rubberized asphalt for various research purposes. The related publications are selected from the collected database and reviewed, in which main revised aging factors and conclusions are summarized in Table 1.

It can be seen from Table 1 that the temperature and duration of

standard thermal aging is widely revised for CRM to trace the composition and rheological changes, investigate the aging mechanisms, and optimize the aging procedure. Based on the results from the rheological tests, the aging duration shows a more dominant role in the thermal aging of CRM in both the short-term aging (Li et al., 2019; Zhao et al., 2022) and long-term aging (Xiao et al., 2009) as compared to aging temperature. This indicates that prolonging the duration of thermal aging is an effective method to comprehensively investigate the aging behavior of CRM, which is also recommended for polymer modified asphalt binder in previous research (Yan et al., 2020). Nevertheless, Liang et al. suggests increasing PAV temperature (Liang et al., 2021) and using a smaller sample pan (32 mm in the diameter) (Liang et al., 2019) for CRM to provide a representative aging condition theoretically, which needs further evidence from field experiments. With the increasing thermal aging duration, the rubber degradation is documented as the main characteristic of CRM (Kataware and Singh, 2019), which is shown by weakened crosslinked network (Cheng et al., 2020), flat phase angle curves (Zhang et al., 2019), and molecular transformations of CRM (Lyu et al., 2021).

4.2. UV aging

As shown in Fig. 2, UV aging of CRM attracts increasing research interests in the recent 5 years, while the standard UV aging procedure for asphalt binder is still missing. Therefore, the sample preparation process and aging conditions indicated in publications in the collected database are summarized and compared in Table 2 and Table 3, respectively.

UV exposure occurs to the asphalt binder during service after the mixture manufacturing and pavement construction. Therefore, UV aging is usually conducted on the short-term aged samples to simulate the field aging conditions (Jin et al., 2019). For the sample preparation of UV aging, the PAV dish (a diameter of 140 ± 0.5 mm) is the most widely used container for CRM according to the collected database due to its availability and standard size. Meanwhile, the thickness of the sample in the PAV dish is suggested to be reduced to 1–1.5 mm for UV aging due to the limited penetration effect of ultraviolet light (Hung et al., 2021). Meanwhile, Zadshir et al. (2018) (Zadshir et al., 2018) proposed an adjusted method based on Practice for Preparation of Test Panels for Accelerated and Outdoor Weathering of Bituminous Coatings (ASTM D1669) (D08 Committee, 2013) to obtain the thinner UV samples (0.25 mm), while the aging effects of the heating treatments during the un-standardized sample preparation have not been evaluated.

Table 1
Revised laboratory thermal aging methods for crumb rubberized asphalt binder.

Standard procedure	Revised factors	Parameters	Main Conclusion
TFOT 163 °C 5 h	Temperature	143, 163, 180, 200, and 220 °C	Increased aging temperature facilitated the desulfurization and degradation of rubber in CRM (Geng et al., 2022; Kataware and Singh, 2019).
	Duration	1, 4, 24, and 48 h	Large molecular size content increases with the increasing aging duration (Lee et al., 2011).
	Temperature	150, 160, 170, 180, and 190 °C	Aging duration has a more significant effect on the deformation recovery ability of CRM (Li et al., 2019).
	Duration	5, 10, 15, and 20 h	The softening points of CRM was not sensitive to aging temperature (Zhao et al., 2022).
	Temperature	150, 163, and 180 °C	
	Duration	5, 10, 15, and 20 h	
RTFOT 163 °C 85 min	Temperature	163, 177, and 195 °C	Higher aging temperature is recommended to simulate the actual aging of CRM (Hemanth Kumar and Suresha, 2019).
	Duration	1, 3, 5 and 7 h	The cross-linked network of CRM is stable within 3 h aging, and then gradually weakened until broke (Cheng et al., 2020; Li et al., 2020).
		70, 85, 100, 115, and 240 min	The high failure temperature of CRM decreased with the increasing aging duration (Lee et al., 2008).
	Temperature	163, 170, 177, 184, and 191 °C	Three main molecular transformations (medium molecular to large molecular, large molecular to small molecular, small molecular to medium molecular and large molecular) appear alternately with the increasing aging duration (Ye et al., 2020).
	Duration	85, 145, 205, 265, 325, and 385 min	
PAV 100 °C 20 h	Duration	20, 40, and 80 h (Ye et al., 2019)	Increased aging duration decreased the low-temperature performance of CRM and gradually separated rubber from the base asphalt (Chang et al., 2020).
		20, 40, and 60 h	The phase angle curves of CRM become flat after 40 h aging (Zhang et al., 2019).
		20 and 40 h	The loss of saturates is observed in the early-stage aging of CRM, followed by the agglomeration of aromatics (Lyu et al., 2021).
	Temperature	90 and 100 °C	Carbonyl component shows a consistent increasing trend throughout the long-term aging procedures (Liang et al., 2019).
	Duration	20, 40, and 60 h	
	Temperature	65, 80, 100, and 110 °C	Increasing aging duration causes an increase in creep stiffness and a decrease in m-values (Xiao et al., 2009).

“Heat-casting method” is also adopted by Hung et al. (2021) (Hung et al., 2021) for the UV aging sample preparation of CRM, which is only suitable for small quality of samples (≤ 6 g).

Ultraviolet light source, temperature, and duration of laboratory UV aging are described in detail in each related publications, indicating their dominant aging effects on samples. The wavelength of UV lamps mainly concentrated around 300 nm, which is similar to the wavelengths of ultraviolet light from the sunlight (Liu et al., 2021). The UV aging temperatures ranges from 25 °C to 90 °C in different publications,

Table 2
Sample preparation for UV aging of crumb rubberized asphalt binder.

Sample container	Weight/Thickness	Pretreatment
Silicone rubber dish (Hung et al., 2021)	6 g sample	135 °C heating
PAV dish	50 g sample (Jin et al., 2019; Liu et al., 2021) 1 mm thick (Pang et al., 2014) 3 mm thick (Zhang et al., 2018) 1.5 mm thick (Qian et al., 2020)	TFOT
Aging tray (Osmari et al., 2019)	0.7 mm thick	–
Aluminum plates (Zadshir et al., 2020)	–	135 °C heating

Table 3
UV aging methods for crumb rubberized asphalt binder.

Methods/Devices	UV source and parameters	Distance	Temperature	Duration
QUV accelerated weathering machine	UVB-313 EL, 0.71 W/m ² /nm at 313 nm (Hung et al., 2021)	10 cm	65 °C	100 and 200 h
	UVA-340 nm bulbs with the wavelength of 95–365 nm	–	–	869 h (Jamal et al., 2021)
		–	45 °C	100 h (Zadshir et al., 2020)
UV aging chamber	UV lamp with 3.18 W/m ² (Jin et al., 2019)	–	–	336 h
	300 W UV lamp with the wavelength of 280 nm–400 nm (Liu et al., 2021)	–	60 °C	3, 6, and 9 days
	UV lamp, 26.5 W/m ² with the wavelength of 360 nm (Pang et al., 2014)	–	60 °C	10 days
	UV lamp, 14.2 mW/cm ² (Qian et al., 2020)	–	25 °C	7 days
	1 kW high pressure mercury lamp (L. L. Wang et al., 2021)	35 cm	40–50 °C	200, 600, and 1000 h
	500 w UV lamp (Zhang et al., 2018; Zhu et al., 2017)	–	60 °C	6 days
Suntest CPS equipment (Osmari et al., 2019)	Xenon lamp, 70 mW/cm ²	–	90 °C	120 h
Artificial intense ultraviolet radiation environment box (Wu, 2017)	High-pressure mercury lamps	–	50 °C	50, 100, 200, and 300 h

which still need further comparison research between laboratory and field to be furtherly optimized. Meanwhile, the UV aging duration of CRM also varies from 50 h to 1000 h, which depends on the research purpose and the simulated weather indicated in the publications. For investigating the UV aging mechanisms, a 50–200 h aging duration is widely adopted (Hung et al., 2021; Wu, 2017; Zadshir et al., 2020), while a longer aging duration is usually used to simulate the aging behavior of CRM exposed to a certain level of ultraviolet light (Jamal

et al., 2021). Zadshir et al. (2020) (Zadshir et al., 2020) introduced the 4h rest period during UV aging to simulate the nighttime, while no significant difference was observed with this modification. Nevertheless, the distance between the lamps to samples is an easily ignored factor of the UV aging procedure in the existing publications, which is proven to determine the irradiation intensity on the surface of samples (Y. Li et al., 2022).

4.3. Weathering aging

In addition to oxygen, heat, ultraviolet (UV) light, moisture is another important factor to determine the aging behavior of CRM (D. D. Hu et al., 2021). The collected database shows that the effect of water on aging of CRM is investigated by a modified PAV test, which includes one-third PAV aging period with water and the following two third PAV aging period without water (Xiao et al., 2012, 2017). As compared to this modified PAV test, the accelerated weathering aging test with adjustable conditions may simulate in situ aging of CRM closer (M. M. Hu et al., 2021). Various radiation intensities (0, 500, 1000 w/m²) were conducted on the asphalt samples with various thickness (100, 300, 500, and 1000 μm) at the target temperature (70 °C) and humidity (70%), to simulate aging conditions in different temperature-climate regions as well as investigate the effect of solar radiation intensity on the aging depth distribution. The study of Mirwald et al. (2020) (Mirwald et al., 2020) proposed the Viennese Binder Aging method, that incorporates two ROS (O₃ and NO_x) into the aging atmosphere, which realistically simulates the field aging conditions. However, there are still several factors (vehicle loading and acidic components in the atmosphere to name a few) affecting the aging of CRM, which cannot be fully considered and simulated in the laboratory aging experiments. Therefore, several studies exposed the samples to the field environment to obtain the weathering aged CRM, in which the aging duration varies from 9 months to 48 months (Wang et al., 2016; Y. Y. Wang et al., 2021). The sample for field aging usually obtained by coating an aluminum plate with the CRM and then placing it in an open field (S. S. Wang et al., 2017). Nevertheless, weather aging usually requires a long testing period, and the results are only referential for the same climate zones. Meanwhile, the correlation between the laboratory aging and field aging of CRM needs to be comprehensively investigated for the following research, which provides the guidance for the optimization of aging procedure for CRM.

5. Chemical-microstructural characterization

As the byproduct of petroleum distillation processes, asphalt binder is a complex mixture of tens of thousands of different chemical components, the interactions of which form various microstructures (Pfeiffer and Saal, 1940). The chemical components and microstructures are correlated to the bulk properties of asphalt binder (Yu et al., 2019), while also changed during the aging process (Mikhailenko et al., 2019). Therefore, the characterizations of chemical components and microstructures are effective to unravel the aging mechanisms of asphalt binders. With the addition of crumb rubber, the aging of CRM become a more complicated process including the aging of asphalt binders and rubbers alone, as well as the evolution of their interactions (Lyu et al., 2022b). The chemical-microstructural characterizations are widely used to provide insights into the underlying aging mechanisms of CRM.

5.1. Chemical components

5.1.1. Fourier-transform infrared spectroscopy

Fourier-transform infrared spectroscopy (FTIR) and gel permeation chromatography (GPC) are the high-frequency chemical characterization techniques used for the characterization of aging of CRM asphalt based on the collected documents. As a reliable and convenient characterization method, FTIR can identify the molecular fingerprints from

different chemical structures of asphalt binders by detecting the transmitted or reflected infrared signals (Hofko et al., 2018; Poulikakos et al., 2019). Therefore, the components of CRM were widely documented by FTIR after different aging procedures to compare the chemical evolution induced by various environmental factors and the detailed comparisons were summarized in Table 4. The continuous increase in sulfoxide and carbonyl is the most significant aging-related change for asphalt binder (Poulikakos et al., 2019), which is also observed in thermal aged and UV aged CRM (Lyu et al., 2021). Specifically, Li et al. (2020) (Li et al., 2020) showed an initial decrease in the sulfoxide of CRM during the short-term thermal aging (0–1 h) followed by an increase (3–5 h) and disappear (7 h), which is speculated to be related to the chemical bonds transformed in sulfide within crumb rubber. Meanwhile, the characteristic peaks of natural rubber and synthetic rubber (724 cm⁻¹, 810 cm⁻¹, and 967 cm⁻¹) are also reported to decrease in CRM during thermal aging (Geng et al., 2022), which is attributed to the chain scission reaction of crumb rubber. This indicates that the degradation of crumb rubber is also a major phenomenon during the aging process of CRM, synergistically affecting the chemical changes with the aging of asphalt binder (Geng et al., 2022; Nivitha et al., 2016). A significant divarication is observed in the ratio change of aromaticity components in CRM during thermal aging, which may be attributed to the different selected peak wave-numbers (1596 cm⁻¹, 1604 cm⁻¹, or 1535–1625 cm⁻¹) and calculation methods (peak height or area) (Chang et al., 2020; Cheng et al., 2020; Li et al., 2020). Considering the limit of FTIR, Siddiqui et al. (1999) adopted Nuclear Magnetic Resonance to measure the aromatization content of base asphalt binder during TFOT and PAV aged binder, which may be also useful to unravel the evolution of aromatization in CRM. Meanwhile, several researchers claim that the nano-silica component from crumb rubber causes a strong peak at 1100 cm⁻¹ and enhances the thermal aging resistance of CRM (Tang et al., 2019; Wang and Huang, 2021), while little attention is focused on the change in silica-related peaks of CRM during the aging process.

5.1.2. Gel permeation chromatography

GPC refers to a liquid chromatography technology, which can separate different polymer compositions based on their molecular hydrodynamic volumes (Baek et al., 2009). Specifically, the larger molecules quickly elute in the connected columns and form the peak at a shorter elution time or retention time of GPC chromatograms. Before injecting into the GPC device, CRM samples are usually dissolved by tetrahydrofuran solution and preserved for 24 h to achieve full dissolution (Li et al., 2017). Ma et al. (2020) proposed that 24 h preservation should be extended to 72 h for CRM due to the existing cross-linked polymers components. However, the subsequent research shows that the extended preservation may result in the extraction of soluble components from crumb rubber (Ma et al., 2020), which may be confused with the soluble rubber compositions in asphalt binder. Therefore, the concentration of CRM solution and preservation duration should be consistent in each test to ensure reproducibility in the following research. The raw data from the GPC test should also be calibrated and normalized to eliminate the potential operation deviation.

For the GPC chromatograms, the areas between the X axis and normalized refractive index curve at different elution periods are the common indicators for analyzing asphalt binder (Ma et al., 2021). The area is usually divided into 13 slices with equal interval of retention time, in which the 1–5, 6–9, and 10–13 slices refers to the large molecular size (LMS), medium molecular size (MMS), and small molecular size (SMS), respectively (Putman and Amirkhani, 2010). Several researchers show that a continuously increasing LMS appears in the GPC as a result of the thermal aging of CRM (TFOT and PAV), which is consistent with the aging behavior base asphalt binder (Kim et al., 2006; Ma et al., 2020; Zhou et al., 2020). This can be easily understood by the aging-induced transformation of aromatics to asphaltenes (Lee et al., 2011). However, the research of Lee et al. (2011) (Lee et al., 2011) and Cheng et al. (2020) (Cheng et al., 2020) report an initial increase

Table 4

Functional group changes of crumb rubberized asphalt binder during aging.

Aging condition	TFOT			PAV		UV	
	163 °C 5 h (Ma et al., 2020; Nivitha et al., 2016; Zhang and Hu, 2015)	180–220 °C 5 h (Geng et al., 2022)	120 °C 16–185 h (Nivitha et al., 2016)	100 °C 10 h (Chang et al., 2020; Geng et al., 2022)	100 °C 20 h (Chang et al., 2020; Geng et al., 2022; Ma et al., 2020; Nivitha et al., 2016)	100 °C 50–150 h (Lyu et al., 2021)	45 °C 100 h (Zadshir et al., 2020)
724/810/967 Rubber		↓		↓	↓		
967.8 CH=CH	↑						
1030 Sulfoxide	↑				↑		
1376 Aliphatic bond							↑
1535–1625 Aromaticity	↓		↓		↑		
1604/1596 C=C				↓	↓		
1700 Carbonyl	↑				↑		↑
2968 CH ₃						↑	
3200–3500 O–H							↑

*The first column shows the wavenumbers (cm⁻¹) and corresponding components. Upward and downward arrows refer to increase and decrease, respectively. Multiple arrows indicate trends over aging time.

followed by a decrease in LMS of CRM during thermal aging. This may be attributed to the multiple aging-induced evolutions within CRM: the first evolution is the agglomeration of light components to increase the molecular size; the second one is the degradation of rubber components to decrease the molecular size (Ma et al., 2020). As for UV aging, the research of Lyu et al. (2021) (Lyu et al., 2021) shows a significant decrease in the LMS of CRM, indicating that the degradation of crumb rubber plays a dominant role in UV-induced aging. It's remarkable that the decreasing LMS does not indicate less agglomeration of CRM, which can be characterized by the spectrophotometry technique (Hou et al., 2018).

5.2. Microstructure

Characteristic microstructures of asphalt binders are reported to largely determine the bulk rheological and thermal properties (Yu et al., 2019). Scanning electron microscope (SEM) and atomic force microscopy (AFM) is the most widely used microstructure characterizations used that provide insights into the aging mechanism of CRM. By the observation of SEM, the addition of crumb rubber is proven to mitigate the formation of the crack in the surface of asphalt binders during thermal aging (G. Wang et al., 2020) and UV (Zadshir et al., 2020). The aging-induced changes are also observed in the interaction area between crumb rubber and asphalt binder within CRM. Specifically, the stripping behavior of rubber particles from the asphalt binder is the main aging evolution of CRM during the thermal aging (RTFO and PAV) (Kumar et al., 2020). Meanwhile, the size of rubber particles significantly increases during the 20 h PAV (Zhou et al., 2021) followed by a decrease with the longer aging period (Chang et al., 2020), which may be attributed to the swelling and degradation of rubber particles of CRM during the aging process. Mikhailenko et al. (2019) (Mikhailenko et al., 2019) used Environmental Scanning Electron Microscopy to trace the evolution of fibril structures within the base asphalt binder as a result of aging, showing the denser fibril structures after RTFOT and PAV. Using this technique on CRM shows that crumb rubber interrupts the single-phase fibril structure of asphalt binder, demonstrating the clear interface surrounding rubber particles (Lyu et al., 2022a). This remains a research gap on the aging-induced evolution of fibril structure and rubber-asphalt interface within CRM.

AFM is another method used to investigate the micro-scale properties of asphalt binder due to its nanoscale precision (Hung and Fini, 2020). Catana (bee-structure), peri (surrounding area), and para domains are the three dominant domains in the AFM images of asphalt binders (Masson et al., 2006). The study of Kim et al. (2017) (Kim et al., 2017) shows that the number of bee-structure of CRM decreases during thermal aging, which is contrary to the aging behavior of base asphalt. The similar results were also reported by Lv et al. (2021) (Lv et al., 2021), which may be attributed to the inhibiting effect of rubber to the crystallization in the surface of asphalt binders. Nevertheless, the opposite phenomenon was shown by Huang and Pauli (2008) (Huang and Pauli, 2008) that thermal aging results in fewer bee-structures with larger sizes within CRM. The study of Lyu et al. (2023) (Lyu et al., 2023) prepared 5 repetitions to trace the aging-induced domain changes of CRM quantitatively, showing the increased numbers and decreased size of bee-structures within CRM after aging as shown in Fig. 3. The divided opinion may be attributed to the different raw materials and modification methods. The AFM investigation in the aging of CRM mainly limits in the thermal aging process (RTFO and PAV), which remains a research niche in the UV-induced surface evolution. Meanwhile, more advanced AFM-based nanoscale characterizations enable to observe the micro-mechanics and microchemical properties, which may benefit understanding the underlying aging mechanisms of CRM. The study of Rodríguez-Fernández et al. (2020) (Rodríguez-Fernández et al., 2020) used atomic force microscopy-based infrared spectroscopy (AFM-IR) to observed the distribution of carbonyl and sulfoxide bonds in CRM as shown in Fig. 4, which can trace the generation of aging-related compositions during aging. Lyu et al. (2023) (Lyu et al., 2023) recently introduced the Peak-force tapping quantitative nanomechanical test to investigate the micromechanical features of CRM after aging, tracking the evolution of surface modulus and adhesion at each domain. In addition, Li et al. (2022) (DN Li et al., 2022) investigated the micro-structure evolution of CRM during thermal aging from the perspective of multiphase structure as shown in Fig. 5, unraveling the chemical and mechanical changes of different microstructures within CRM during aging.

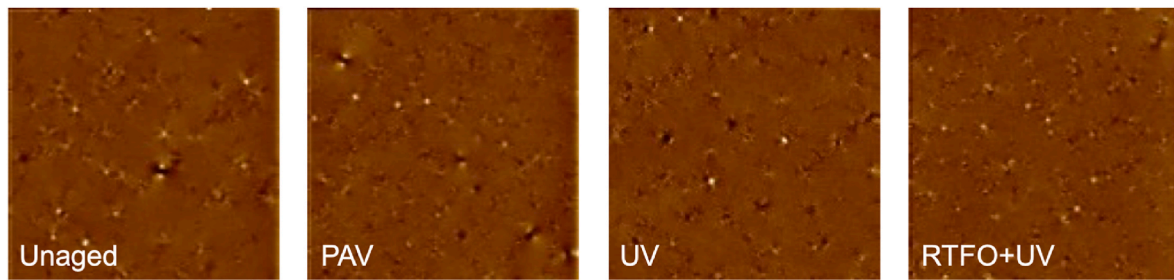


Fig. 3. AFM topographical images of CRM before and after PAV, UV aging, and RTFO + UV aging, showing the increased numbers and decreased sizes of bee-structures (Lyu et al., 2023).

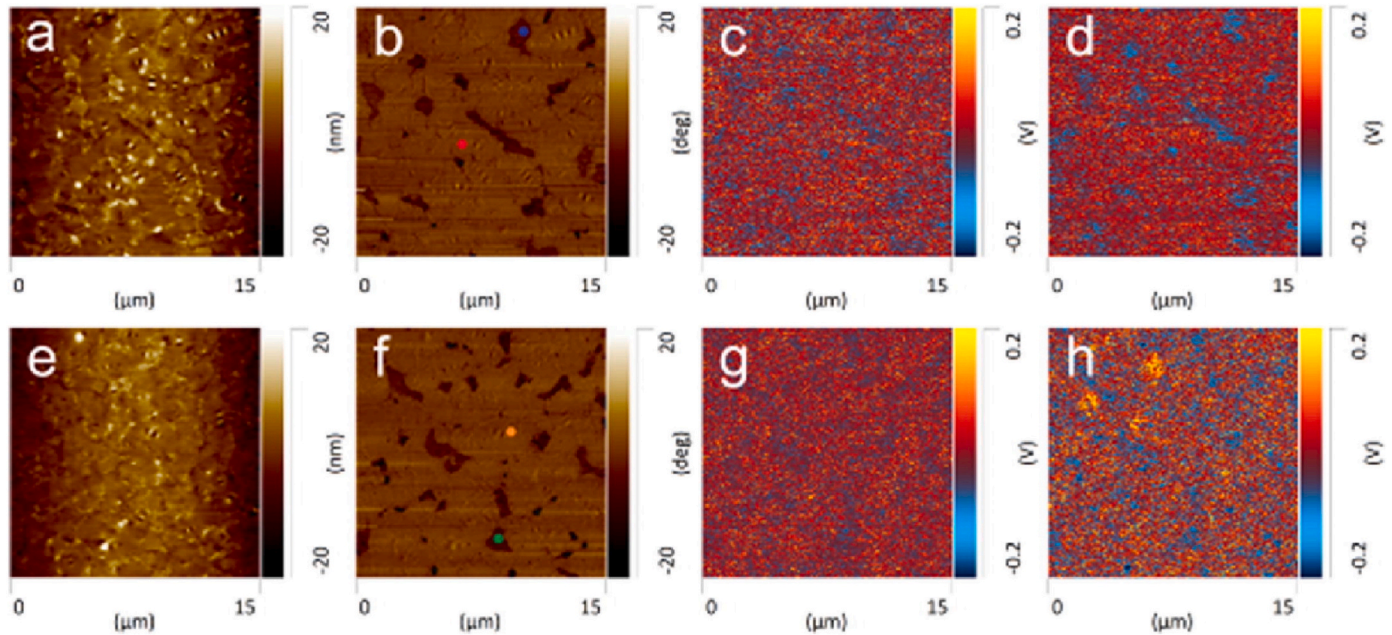


Fig. 4. AFM-IR results for CRM modified for 30 min (a–d) and 180 min (e–h). (a, e) Topography; (b, f) Phase; (c, g) IR maps of sulfoxide groups at the wavenumber of 1030 cm^{-1} ; (d, h) IR maps of carboxyl functional groups 1700 cm^{-1} (Rodríguez-Fernández et al., 2020).

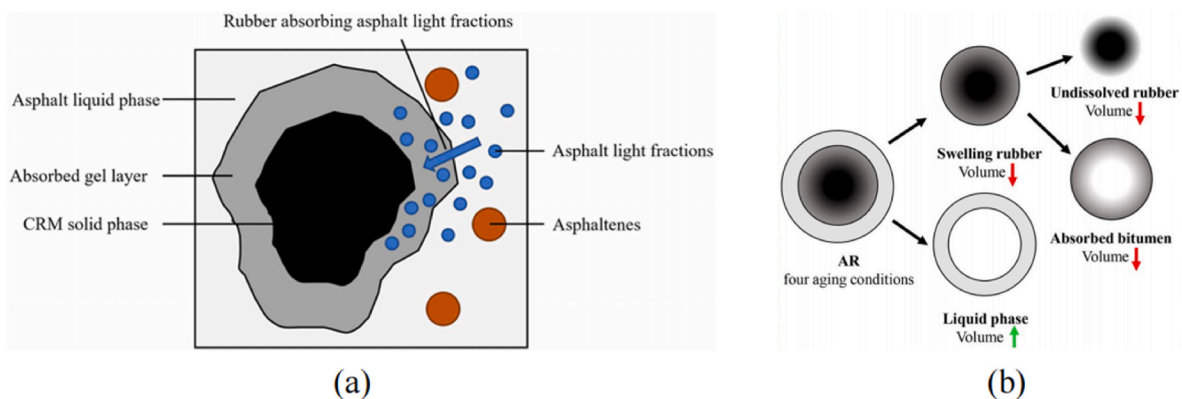


Fig. 5. (a) Schematic of multiphase structure within CRM (Li et al., 2021); (b) The volume evolution of phases within CRM during thermal aging (DN Li et al., 2022).

6. Rheological-mechanical properties

Asphalt binder as a viscoelastic material has a range of behaviors ranging from sol (Newtonian dominated) to gel (non-Newtonian dominated), affected by its colloidal system as well as the environmental factors (Sybilski, 1993). The ratio of the elastic and viscous components in asphalt binders also determines their rheological-mechanical

properties subjected to different temperatures and loadings. Meanwhile, the rheological properties of asphalt binder are proven to be susceptible to environmental aging, which may subsequently cause the degradation of pavement structures (Petersen, 2009). Therefore, several rheological-mechanical indicators are used to understand the rheological-mechanical evolution of CRM as a result of the aging processes, including high and low temperature properties, fatigue

properties, and moisture damage resistance.

6.1. High-temperature properties

The viscosity and deformation resistance are the two major rheological properties of CRM, which are widely compared before and after aging. The viscosity of CRM is usually measured after short-term thermal aging to evaluate its blending workability (Wang et al., 2012b). As mentioned earlier, aging results in the loss of aromatic oil and light fractions, thereby increasing the viscosity of asphalt binders. Nevertheless, Wang et al. (2012a) (Wang et al., 2012b) reported a significant decrease in the viscosity of CRM after short-term thermal aging, especially at the high rubber content ($\geq 25\%$). As for the deformation resistance, high failure temperature, phase angle (δ), complex modulus (G^*), rutting resistance parameter ($G^*/\sin \delta$), and non-recoverable compliance are used to track the evolution of CRM during aging as summarized in Table 5. It is shown that some CRMs demonstrate weakened high-temperature deformation resistance, which is contrary to the base asphalt binder. Ye et al. (2019) (Ye et al., 2019) proposed that the weakened high temperature properties are mainly attributed to the aging-induced degradation of rubber. This reduction is confirmed by the study of Wang et al. (2012a) (Wang et al., 2012a), showing that the deterioration of high temperature properties mainly occurs in the CRM with high rubber content ($\geq 15\%$). This indicates a pair of competitive evolution tendencies in CMR during aging: the base asphalt binder tends to be stiffer, while degradation of rubber may decrease the stiffness of CRM. The high temperature properties are finally determined by the dominant mechanism, which depends on the rubber content, aging duration, and other potential factors. Generally, aged CRMs always demonstrate better high-temperature properties than the base asphalt binder (Chen et al., 2018; Lee et al., 2008), indicating that high-temperature properties of aged CRMs is not the main drawbacks to limit the durability of asphalt pavements.

6.2. Low temperature properties

Aging-induced embrittlement of asphalt binder increases the cracking risk at low temperatures. The Bending beam rheometer (BBR) test (ASTM D6648-08, 2016) is widely used to evaluate the low temperature properties of aged CRM, in which a lower creep stiffness and higher creep slope (m -value) indicate a better low temperature cracking resistance. Studies show that thermal aging and UV aging always results in a weakened low temperature behavior of CRM but it is significantly better than aged base asphalt binder and styrene-butadiene-styrene modified asphalt binder (Wang et al., 2015), indicating the low-temperature properties of aged CRMs are not of primary concern. Ye

et al. (2019) (Ye et al., 2019) added the crumb rubber to the styrene-butadiene-styrene modified asphalt binder, thereby mitigating its degradation in low temperature properties after thermal aging. The enhanced cracking resistance is mainly attributed to the blocking effect of rubber particles to the generation of cracks (T. T. Wang et al., 2017). Meanwhile, microwaving (Liang et al., 2017), trans-polyoctenamer, and twin-screw extrusion (Rasool et al., 2018) treatments on crumb rubber are shown to furtherly enhance the cracking resistance of aged CRM by facilitating the interaction between rubber particles and asphalt binder.

6.3. Fatigue properties

Fatigue properties of CRM are reported to continuously decrease with the longer aging duration, which is consistent with the performance of the base asphalt binder (Xiao et al., 2017). Nevertheless, the comparison in fatigue properties of base asphalt binder, styrene-butadiene-styrene modified asphalt binder, and CRM during aging demonstrates the different results due to the selection of fatigue indicators. Wang et al. (2015) (Wang et al., 2015) used the fatigue factor ($G^*\sin \delta$) to compare the fatigue properties of aged CRM, base asphalt binder, and styrene-butadiene-styrene modified asphalt binder, showing a better fatigue performance with the addition of crumb rubber than styrene-butadiene-styrene after aging. similar results are also reported in the studies of Yousefi Kebria et al. (2015) (Yousefi Kebria et al., 2015), Xiao et al. (2017) (Xiao et al., 2017), and Zhang et al. (2020) (Zhang et al., 2020) by the same fatigue indicator. Meanwhile, fatigue life obtained using linear amplitude sweep testing (TP101, 2012) was also adopted to evaluate the fatigue properties of aged CRM, showing that the fatigue life of aged CRM is lower than styrene-butadiene-styrene modified asphalt binder [53] and even the base asphalt binder (Mu et al., 2020). Those two indicators were compared in the study of Wang et al. (2022) (Wang et al., 2022), showing a better correction between fatigue life from linear amplitude sweep testing to the fatigue properties of CRM before and after aging. Furthermore, a recent study (Kumar et al., 2020) discussed five widely-used binder fatigue test procedures and recommended dynamic shear rheometer-based elastic recovery test and binder yield energy test to evaluate the fatigue properties of polymer-modified asphalt binder, which is lacking in the investigations of aged CRM till now. The future investigation on the rheological properties of aged CRM is recommended to focus on studying fatigue properties using dynamic shear rheometer-based elastic recovery test and binder yield energy test, thereby accurately evaluating the fatigue resistance of aged CRM as compared to the base asphalt binder.

6.4. Moisture damage

The addition of crumb rubber has been shown to weaken the moisture resistance of asphalt binders (Kim et al., 2014; Pérez and Pasandín, 2017; Shirini and Imaninasab, 2016), while most studies on the moisture resistance of aged CRM adopted the mixture level experiments. The existing moisture tests as the bitumen bond strength test are dominated by the bulk asphalt properties rather than the interfacial properties. This brings difficulties to unravel the underlying failure mechanisms at the asphalt-aggregates interface under wet conditions. A recent work (Oldham et al., 2022) proposed a novel interfacial moisture resistance test, the moisture-induced shear thinning index, showing good agreement with other laboratory moisture tests as well as providing more insights in the interfacial failure mechanisms. Future research should focus on the degradation in the moisture resistance of CRM during aging at the binder level, thereby providing more insights into the aging-induced moisture damage behavior. Studies have reported progressive aging in neat asphalt increases asphalt interfacial binding to siliceous surfaces (Rajib et al., 2021). However, latter phenomenon in presence of CRM needs to be further examined.

Table 5
Deformation resistance of aged CRM.

Aging procedure	Indictors	Results
RTFO	High failure temperature	- Increased or decreased depend on the binder sources (Akisetty et al., 2009). - Decreased in CRM with high rubber content ($\geq 15\%$) (Wang et al., 2012a). - Increased and subsequently decreased with longer aging duration (100 min) (Lee et al., 2008).
	$G^*/\sin \delta$	- Increased (Wang et al., 2012a). - Ordinary CRM asphalt decreased but desulfurized rubber asphalt increased (J. J. Wang et al., 2020).
	Phase angle	- Decreased (Chen et al., 2019)
TFOT + PAV	Non-recoverable Compliance	- Severely increased (Ye et al., 2019).
RTFO + PAV	G^* and δ	- Increased G^* and decreased δ (Chen et al., 2020).

7. Environmental Product Declaration

Environmental concerns are attracting increasing interest in the pavement industry. Almost all publications on CRM claim that there are environmental benefits for use of crumb rubber from waste tires in asphalt, while very few researchers systematically quantified the environmental impacts of CRM. As a standardized method, life cycle assessment (LCA) is becoming a popular method to evaluate the sustainability of asphalt pavements from raw materials to use phase and end-of-life (Piao et al., 2022). The comparable quantified results from the LCA analysis rely on the estimated service life of asphalt pavements, which is largely determined by the aging properties of asphalt binders. The abovementioned discussions show the relatively enhanced rheological-mechanical properties of CRM after aging, as compared to the base asphalt binder as well as the polymer modified asphalt binder. Therefore, the Environmental Product Declaration (Del Borghi, 2013) is conducted on CRM to communicate the environmental performance. National Asphalt Pavement Association investigated industry-average life cycle inventory for North American industry conditions, showing a decrease of 19% and 5% in the global warming potential and non-renewable primary energy resources when comparing CRM to polymer modified asphalt binder (Wildnauer et al., 2019). The study of Farina et al. (2017) (Farina et al., 2017) based on the database of European Bitumen Association, also indicates that the incorporation of CRM into asphalt pavements decreased the life cycle environmental burdens by 23%.

Peeyush et al. (2020) (Khare et al., 2020) reports that asphalt-related secondary organic aerosol precursor emissions from road and roofing exceed those from motor vehicles on urban scales. However, the existing LCA on CRM ignored the potential atmospheric emissions and toxic leaching of the asphalt pavement during environmental aging, which may result in the environmental burden. Remarkably, studies already show that the incorporation of crumb rubber into asphalt binder results in several times higher pollutant emissions during production as compared to the base asphalt binder (Bueno et al., 2021; H. Li et al., 2023; Xie et al., 2023; Yang et al., 2019). The studies of Tang et al. (2022) (Tang et al., 2022) and Li et al. (2023) (H. Li et al., 2023) also indicate that the addition of crumb rubber in asphalt binder may even cause several new hazardous emissions (benzothiazole, N-cyclohexylcyclohexanimine, 2,2,4-trimethyl-1,2-dihydroquinone et al.) mainly attributed to additives used in rubber manufacturing. It is reported that bio-modification process can be beneficial to retain some of the volatile compounds (Pahlavan et al., 2023). There have been also studies on retaining emission of asphalt by increasing the intermolecular networks within asphalt (Mousavi et al., 2023a, 2023b). The concerns with emission of CRM during production or during aging, highlight the importance of developing interventions to retain volatile organic compounds in asphalt. Moreover, the study of Piao et al. (2022) (Piao et al., 2022) introduced the leaching tests to dry-process rubberized asphalt pavement to measure the release of polycyclic aromatic hydrocarbons, which should also be conducted on the CRM during aging. An important parameter in the LCA analysis is the local waste management policy and what is done with the waste tires. In some countries such as Switzerland the waste tires are used as fuel mostly in the cement industry and redirecting this waste stream to road construction needs to consider its original route.

Another path to enhance the environmental benefits of CRM is the recycling use of the aged CRM materials to construct the pavement. Despite large number of studies investigated the reclaimed asphalt pavement till now, little attention focused on the reclaimed CRM pavement due to its complex components. The study of Li et al. (2022) (Danning Li et al., 2022) established mobilization indexes based on FTIR and thermal gravimetric analysis to evaluate the mobilization tendencies of rubber and asphalt binders separately. The evaluating results based on mobilization indexes show that the conventional reclaim methods can achieve the simultaneous mobilization of rubber and

asphalt binder in aged CRM. Another recent study (D. Li et al., 2023) proposed a compound rejuvenation scheme that combined the rejuvenator, CRM, and extra crumb rubber modifier, which significantly enhanced the rejuvenation effectiveness of aged CRM.

8. Conclusion and future work

This paper aims to provide a systematic literature review on the aging of CRM based on 560 papers from 1993 to 2022, mainly from the perspectives of aging methods, chemical-microstructural characterization, and rheological-mechanical properties. Widely used CRM aging-related laboratory investigations and evaluation indicators are comprehensively compared and reviewed to unravel the underlying evolution mechanisms of CRM subjected to different aging conditions. The conclusions are drawn as follows:

- (1) Thermal aging effect on CRM still attracts the most research attention, while more studies are gradually focusing on UV aging and weathering aging of CRM. Recent findings demonstrated the complex chemical-physical evolution of CRM aging, which is determined by a combination of multiple factors (humidity, temperature, and solar radiation intensity to name a few).
- (2) Significant decrease in the rubber-related peaks are reported as a characteristic phenomenon in the FTIR spectrum of aged CRM, as well as the common increase in the sulfoxide and carbonyl peaks. As compared to base asphalt, an increase in large-size molecules is followed by an extra decrease in long-term aged CRM, especially in the high-rubber-content CRM. Microstructure characterization shows that rubber mitigates the formation of aging-induced microcracks and thermal aging decreased the number of bee structures in nanometer scale.
- (3) Rheological-mechanical investigations show that aging has various effects on the high-temperature properties of CRM depending on the modification procedures and aging conditions, while aged CRM demonstrates superior low-temperature properties as compared to base asphalt and styrene-butadiene-styrene modified asphalt. Degradation in fatigue properties of CRM during aging should attract more concerns, thereby enhancing the durability of asphalt pavements.
- (4) Recent studies show that the aging evolution of CRM includes three major mechanisms: asphalt aging, rubber degradation, and components exchange between rubber and asphalt. All three mechanisms have various contributions to the overall chemical-mechanical properties of aged CRM, while the dominant mechanism is determined by intrinsic properties of CRM (rubber content, particle size, modification methods) and aging conditions.
- (5) Life cycle assessment revealed that substantial environmental benefits of CRM in terms of reduced energy consumption and global warming potential is possible, depending on the local waste management strategy. However, it is important to also consider potential environmental impacts during the service life, such as atmospheric emissions and toxic leaching.

In future studies, three aspects need further investigation: (1) Developing laboratory aging methods to simulate the in-field aging environment of CRM and comprehensively investigating UV-induced aging behavior; (2) Tracking components exchange within CRM during aging, which may play a dominant role in microstructures and bulk properties of aged CRM; (3) Assessing the environmental impacts of CRM during aging, including atmospheric emissions and toxic leaching.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Lily Poulikakos reports financial support, administrative support, article publishing charges, and equipment or supplies were provided by Empa Materials Science and Technology. Lei Lyu reports a relationship with Chang'an University that includes: funding grants and travel reimbursement. Lily Poulikakos has no pending patents. There is no conflict of interest.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2023.140202>.

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